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HEALTH PHYSICS

The Official Journal of the Health Physics Society

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LIST OF CONTENTS

NUMBERS 1/2

R. K. HILLIARD, C. E. LINDEROTH and A. J. SCOTT: Fission product release from overheated uranium—a laboratory study 1
R. A. HARVEY: Some trends and recent health physics instrumentation developments at Hanford . 11
EGILDA D. WITHERELL and RUSSELL F. COWING: Status of state legislation related to ionizing radiation 17
D. G. OTT, W. H. SCHWEITZER, J. A. SAYEG and P. S. HARRIS: Trichloroethylene-saturated water and tetrachloroethylene chemical dosimeter systems—neutron response 20
SOHEI KONDO: Responses of silver-activated phosphate glass to α-, β-, γ-rays and neutrons . . 25
J. L. WOLFSON and J. C. DYMENT: Dosimetric properties of electrets 36
ALAN R. SMITH: A cobalt neutron-flux integrator 40
NATHANIEL F. BARR, MARY B. STARK, JOAN HANDS and JOHN S. LAUGHLIN: Dosimetry with small silver-activated glass rods 48
W. J. BAIR, D. H. WILLARD and L. A. TEMPLE: Plutonium inhalation studies—I. The retention and translocation of inhaled $Pu^{239}O_2$ particles in mice 54
G. W. DOLPHIN and I. S. EVE: Some calculations on the acceptable limits for the inhalation of radiostrontium in an emergency 61
C. L. COMAR, R. H. WASSERMAN and A. R. TWARDOCK: Secretion of calcium and strontium into milk 69
R. W. PERKINS: A large well crystal for the direct measurement of trace amounts of radioisotopes in environmental samples 81
STUART C. BLACK: Low-level polonium and radiolead analysis 87
V. HERRERO CLIMENT: An analysis of the Chicago Synchocyclotron personnel doses over a 6-year period 92
R. E. ALEXANDER: Sea water monitoring following radioactive waste disposal operations . . . 106
Notes:
E. P. RESNER, G. D. SCHMIDT and R. J. VAN TURNER: Health physics for the N.S. *Savannah* . . 114
P. C. AEBERSOLD and J. W. HITCH: Development of safety performance tests for radioisotope sealed sources and devices 117
Letters to the Editor:
W. A. MCADAMS: Note on suggested regulations for radiation exposure 120
D. K. DAVIS: Conversion of a Landsverk Model L-21, pocket dosimeter charger for use on a 110-V a.c. circuit 121
L. ARGIERO, S. MANFREDINI and G. PALMAS: Considerations of radioactive fallout in Livorno (Italy) 121
Health Physics Society Placement Column 124

NUMBERS 3/4

E. E. POCHIN and C. F. BARNABY: The effect of pharmacological doses of non-radioactive iodide on the course of radioiodine uptake by the thyroid 125
C. A. ADAMS and J. A. BONNELL: Administration of stable iodide as a means of reducing thyroid irradiation resulting from inhalation of radioactive iodine 127
A. S. CHHABRA and R. K. HUKKOO: Natural and fallout γ-activity in foodstuffs 151
JOHN Z. HEARON: Body-burden problem 157
RICHARD MOORE: A comparison of HTO in plasma and expired water vapor 161
R. WILSON and J. A. CARRUTHERS: Measurement of bone-marrow dose in a human phantom for Co^{60} γ-rays and low-energy X-rays 171
DONALD C. LAWRENCE: Mixed radiation dosimetry of a plutonium–beryllium neutron source . . 179
K. J. SCHIAGER, T. M. ALLEN, S. B. GERBER, G. E. KINSELLA, F. J. KRUPKA and D. W. REILLY: Improved membrane-filter air-sampling methods for environmental control 185

R. E. Cofield: Radioactivity distributions in the metallurgical processing of thorium 191
Stephen V. Kaye and Paul B. Dunaway: Bioaccumulation of radioactive isotopes by herbivorous small mammals 205
C. R. Richmond, J. E. Furchner and G. A. Trafton: Comparison of predicted and measured equilibrium levels for chronically administered Cs^{137} 219
Notes:
J. M. Brown, Jr., J. F. Thompson and H. L. Andrews: Survival of waste containers at ocean depths 227
J. Bourdrez, J. Girault, G. Wormjer and P. Cohen: Tests carried out on organic cation exchange resins for the decontamination of aqueous radioactive effluents 228
News 229

AUTHOR INDEX

Adams, C. A. p. 127
Aebersold, P. C. p. 117
Alexander, R. E. p. 106
Allen, T. M. p. 185
Andrews, H. L. p. 227
Argiero, L. p. 121
Bair, W. J. p. 54
Barnaby, C. F. p. 125
Barr, N. F. p. 48
Black, S. C. p. 87
Bonnell, J. A. p. 127
Bourdrez, J. p. 228
Brown, Jr., J. M. p. 227
Carruthers, J. A. p. 171
Chhabra, A. S. p. 151
Climent, V. H. p. 92
Cofield, R. E. p. 191
Cohen, P. p. 228
Comar, C. L. p. 69
Cowing, R. F. p. 17
Davis, D. K. p. 121
Dolphin, G. W. p. 61
Dunaway, P. B. p. 205
Dyment, J. C. p. 36
Eve, I. S. p. 61
Furchner, J. E. p. 219
Gerber, S. B. p. 185
Girault, J. p. 228
Hands, J. p. 48
Harris, P. S. p. 20
Harvey, R. A. p. 11
Hearon, J. Z. p. 159
Hilliard, R. K. p. 1
Hitch, J. W. p. 117
Hukkoo, R. K. p. 151
Kaye, S. V. p. 205
Kinsella, G. E. p. 185
Kondo, S. p. 25
Krupka, F. J. p. 185
Laughlin, J. S. p. 48
Lawrence, D. C. p. 179
Linderoth, C. E. p. 1
Manfredini, S. p. 121
McAdams, W. A. p. 120
Moore, R. p. 161
Ott, D. G. p. 20
Palmas, G. p. 121
Perkins, R. W. p. 81
Pochin, E. E. p. 125
Reilly, D. W. p. 185
Resner, E. P. p. 114
Richmond, C. R. p. 219
Sayeg, J. A. p. 20
Schiager, K. J. p. 185
Schmidt, G. D. p. 114
Schweitzer, W. H. p. 20
Scott, A. J. p. 1
Smith, A. R. p. 40
Stark, M. B. p. 48
Temple, L. A. p. 54
Thompson, J. F. p. 227
Trafton, G. A. p. 219
Turner, R. J. van p. 114
Twardock, A. R. p. 69
Wasserman, R. H. p. 69
Wilson, R. p. 171
Willard, D. H. p. 54
Witherell, E. D. p. 17
Wolfson, J. L. p. 36
Wormjer, G. p. 228

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FISSION PRODUCT RELEASE FROM OVERHEATED URANIUM—A LABORATORY STUDY

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Abstract—The nature and amount of radioactive material released during a reactor accident are of importance to reactor hazards evaluations, design of containment facilities and establishment of operating criteria. A laboratory study is currently underway at Hanford to provide experimental data which will be pertinent to analyses of accidental release under a wide range of reactor conditions.

The investigation is designed to determine the effects of temperature, time of heating, atmosphere in which heated, irradiation level and specimen size on release of eight key fission-product elements from irradiated normal uranium.

Preliminary tests investigated the oxidation and ignition behaviour of unirradiated uranium in air and steam. Small uranium cylinders were then irradiated to 2×10^{14} *nvt* and heated out-of-reactor under carefully controlled conditions and the fractions of the fission product isotopes leaving the specimen were determined. Temperatures up to 1440°C for as long as 4 hr in atmospheres of air, steam and helium were measured; release in air was from two to ten times greater than in steam, while in helium the release was very low. Iodine, tellurium and xenon were released in proportion to the amount of uranium oxidized. Cesium and ruthenium were found to be semi-volatile. The release of strontium, barium and zirconium was found to be less than 0.2 per cent under all conditions tested.

Efficiencies of several filter media in retaining volatilized fission products were measured. Tests are currently being performed to determine the influence of high irradiation level and specimen size on the fraction of fission products released from heated uranium.

INTRODUCTION

THE principal danger to the public associated with the operation of nuclear reactors is not that of a nuclear explosion but the possible uncontrolled release of the radioactive fission products which they contain.[1] In attempting to predict the consequences of an accidental release of fission products from a nuclear reactor, theorists are faced with the important question, "What fraction of the fission-product inventory would be released in the event of a reactor accident?". The answer to this question is complicated as it depends heavily on the nature of the accident. Various operating and physical plant characteristics could lead to different amounts of fission-product release, e.g. type of reactor fuel, atmosphere in which fuel heating occurs, and nature of the reactor structure and containment facilities. Because of the limited empirical data available concerning fission-product release, most hazards evaluations are based on what are believed to be pessimistic estimates of release rates.[1]

In the past few years, some experimental data have been offered which show that fission products are retained by molten reactor core materials to a greater extent than previously assumed. Especially notable is the laboratory investigation of PARKER, CREEK and MARTIN at Oak Ridge National Laboratory, who heated

*Operated for the Atomic Energy Commission by the General Electric Company under contract no. AT(45-1)-1350.

various types of reactor fuels in several different atmospheres.[2] Also, Mine Safety Appliance Research Corporation measured the release of fission products from Zircaloy-clad PWR type fuel elements heated in steam,[3] and field release experiments were carried out at the National Reactor Testing Station by several co-operating groups.[4] The results of these and other studies have encouraged the belief that early estimates of fission-product release were unduly high.

At Hanford the need is for an understanding of the basic fission-product release mechanisms so that the type, rate and fraction of radioactive materials released from natural uranium under various conditions can be realistically appraised. Therefore, a laboratory investigation was undertaken to determine whether fission-product release can be predicted if conditions existing at the time of heating are known.

The specific objectives of the investigations were to determine: (1) the fractions of key fission-product isotopes released from irradiated normal uranium as functions of temperature and time at temperature; (2) the comparative release in atmospheres of air, steam and helium; (3) the comparative role of uranium oxidation vs. molecular diffusion and evaporation on the release rate; (4) the tendency for released fission products to condense and deposit on tube walls; and (5) the effectiveness of filter media in retaining released fission products.

The work reported here was performed with uranium irradiated to very low levels. Tests are currently being performed to determine the effect of burn-up on release of fission products. Results of various phases of the investigation have been documented as the work progressed.[5–8] The purpose of this article is to present the overall picture of the laboratory study in its present status.

EXPERIMENTAL PROCEDURE

The general approach to the experimental procedure was to heat small specimens under controlled conditions while scrubbing the outlet gas stream free of released radioactive material. After heating, the evolved material was recovered for analysis, and the specimen residue was dissolved to determine the amount of each fission-product isotope remaining. The sum of the evolved plus the residual material was considered to be equal to the total fission-product content. The fraction of each fission-product isotope released was then the ratio of evolved material to the total. Traps, furnace tube and filters were analyzed individually in order to determine the deposition pattern after release from the specimen.

All experiments were conducted using polished natural uranium cylinders $\frac{1}{4}$ in. in diameter by $\frac{3}{4}$ in. long, weighing 11.5 g. The test specimens were irradiated to tracer levels, about $2.5 \times 10^{14} nvt$ and after a short decay period were exposed to the selected atmosphere of either air, steam, or helium for a predetermined time using the combustion apparatus shown schematically in Fig. 1. The equipment used for the steam experiments utilized a water-cooled condenser and condensate trap at the outlet of the furnace tube. In addition, arrangements were made

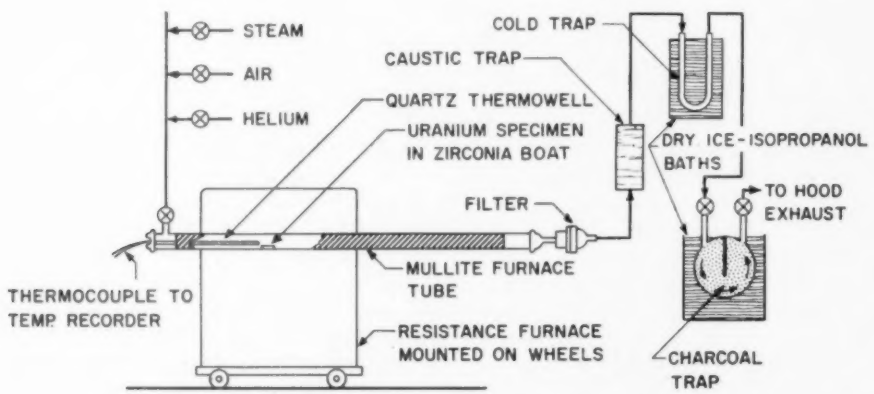


FIG. 1. Flow diagram of fission product release apparatus.

to maintain the inlet and outlet temperature of the steam above 100°C to prevent condensation within the tube.

Air for the release experiments was dried and filtered; helium was dried and passed over copper turnings at 350°C to scavenge residual oxygen. Steam was prepared by boiling distilled water and venting for a minimum of 1 hr to remove dissolved gases.

The experimental procedure was to preheat the furnace to the desired temperature with the irradiated specimen in the cool upstream portion of the combustion tube. With a blanket gas of helium flowing, the furnace, which was mounted on wheels, was rolled into position over the sample. As soon as the specimen temperature approached the desired test condition, helium flow was stopped and the proper atmosphere admitted. The furnace was rolled off the specimen to the downstream end of the combustion tube at the end of the heating period and helium re-introduced concurrently. For the experiments using helium as the atmosphere, the flow of helium was uninterrupted.

The furnace temperature was measured by a platinum vs. platinum–13 per cent rhodium thermocouple located in a quartz well, the tip of which touched the small zirconia boat containing the specimen. A small correction was applied to compensate for the thermal gradient within the furnace tube. Isothermal furnace temperatures were maintained during the tests.

After the specimen had cooled, the furnace tube was removed from the furnace, and the entire residual specimen was transferred to a dissolving apparatus and dissolved in nitric acid or, as was done in several of the tests, the sample was divided into uranium oxide and unoxidized metal and these dissolved separately. During dissolution, the off-gas was scrubbed by a series of traps similar to those used in the heating step.

The furnace tube, connecting lines and off-gas traps were decontaminated to less than 5 per cent of their original value as determined by Geiger–Müller survey counters. The wash solutions were analyzed by conventional radiochemical and gamma spectrometric methods for eight fission-product elements selected for investigation. These were iodine, tellurium, xenon, strontium, cesium, ruthenium, barium and zirconium. They were selected for study on the basis of volatility, biological hazard and abundance in the irradiated fuel.

Since temperature and heating time were two of the primary variables investigated in these experiments, it was considered necessary also to obtain oxidation data. This was done in a series of preliminary tests using unirradiated uranium to determine oxidation rates over the temperature range of interest. Some uranium oxidation data were also obtained as part of the fission-product release experiment. The temperature range investigated in both the fission-product release and oxidation experiments was 405–1440°C with heating times of from 2.5 min to nearly 4 hr. The flow rate of the gas over the test specimens was the same in all cases, 2300 cm^3/min at standard conditions. All tests were made at atmospheric pressure.

RESULTS

A review of the experimentally measured fractional release of radioisotopes for the tests performed in air and steam provides a means for grouping the isotopes roughly in the order of their decreasing volatility, that is:

Volatile	Iodine, Tellurium, Xenon
Semivolatile	Cesium, Ruthenium
Nonvolatile	Strontium, Zirconium, Barium

When ranking the results obtained from the tests in helium atmosphere, cesium and ruthenium could equally well be included in the nonvolatile category. As will be seen later the above arbitrary grouping of volatilities is merely an aid for interpreting the results of these experiments. In all discussions concerning these data, release is defined as that fraction of the radioisotope which escaped from the heated specimen.

Release of semi- and non-volatile elements

Release data for the semi- and nonvolatile elements are presented in Table 1. Because of the small fractions released it was not possible to correlate the release of Ru^{103}, $Cs^{136-137}$, Sr^{89-90}, Ba^{140} and Zr^{95} against the variables of temperature, time of heating or atmosphere except to a limited degree. In general, release of semi- and non-volatiles was highest in air, with steam next in order, and helium providing the least opportunity for escape. It should be noted that the release of cesium and ruthenium is substantially higher than that of the nonvolatile

Table 1. *Release of semi- and nonvolatile fission products in air, steam and helium atmospheres*

Atmosphere		Per cent released from specimen*				
		Ru^{103}	$Cs^{136-137}$	Sr^{89-90}	Ba^{140}	Zr^{95}
Air	Minimum	0.04	0.2	0.001	0.004	0.003
	Maximum	16	6	0.1	0.16	0.07
	Average	3	1	0.03	0.06	0.03
Steam	Minimum	0.007	0.3	0.006	0.005	0.01
	Maximum	0.9	1.3	0.03	0.05	0.1
	Average	0.4	0.7	0.02	0.02	0.04
Helium	Minimum	0.008	0.04	0.008	0.01	0.007
	Maximum	0.13	0.17	0.18	0.23	0.45
	Average	0.05	0.08	0.06	0.07	0.14

* Results are from experiments ranging in temperature from 425°C to 1440°C and in time from 2.5 min to 4 hr.

components, while still low compared to iodine, tellurium and xenon.

Ruthenium behaved in an anomalous fashion for the tests in air. It was released at a higher rate after all the uranium had been converted to oxide. Its highest release was observed at the relatively low temperature of 535°C. The release of cesium was lower than would be expected from a consideration of its relatively low boiling point. Its release was only slightly time and temperature dependent.

It is encouraging to find that the release of strontium was very low in all three atmospheres —varying from 0.001 to 0.18 per cent in all the tests made. Barium and zirconium were released to about the same extent as strontium. The release of these three elements did not appear to depend on time of heating, but was slightly temperature dependent.

Release of volatile elements

A table of ranges of release percentages for "volatile" fission-product elements, such as listed in Table 1 for semi- and nonvolatile elements, is not presented here. Tabular results would not display the correlation with time and temperature observed for the volatile elements. Instead, the results are presented graphically as functions of time and temperature.

The time that the uranium specimens were exposed to an elevated temperature proved to be an important variable in the release of volatile fission products. The release rate was rapid initially but decreased as the heating period progressed. Fig. 2 shows this effect graphically for release of xenon, iodine and tellurium in atmospheres of air and steam at a temperature of 1215°C. The effect of time of heating cannot be fully appreciated without concurrently considering the oxidation reaction which is discussed in detail in a following section. It is sufficient to point out here that the uranium specimen was completely oxidized at 1215°C in air in 48 min, while in steam at the same conditions only 24 per cent of the metal was oxidized. Fig. 2 illustrates that release in air proceeds very slowly after the uranium is completely oxidized. Oxidation rates in steam were so much lower than in air that no test was conducted sufficiently long to oxidize the uranium completely.

As would be expected from kinetic and vapor pressure considerations, temperature had a profound effect on the fraction of volatile fission products released. Fig. 3 is a graph of the release of xenon, iodine and tellurium in air and steam atmospheres as a function of furnace temperature at a constant heating time of 24 min. Once again it is seen that release in air is considerably greater than in steam at all temperatures investigated. Release in helium

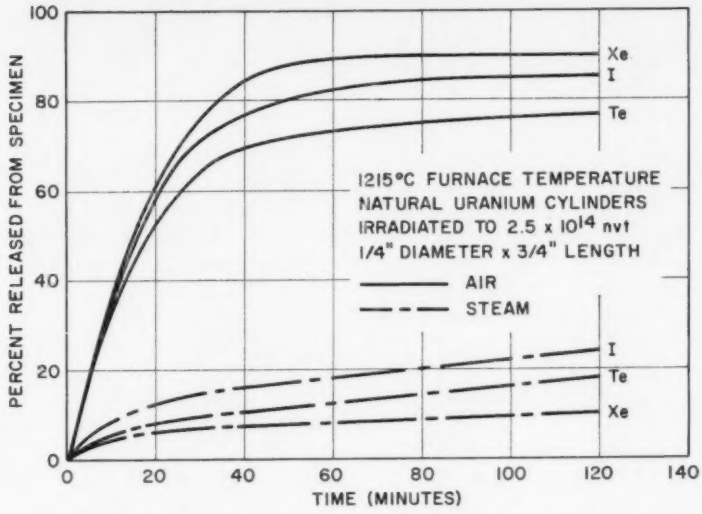


FIG. 2. Effect of time on release of volatile fission products.

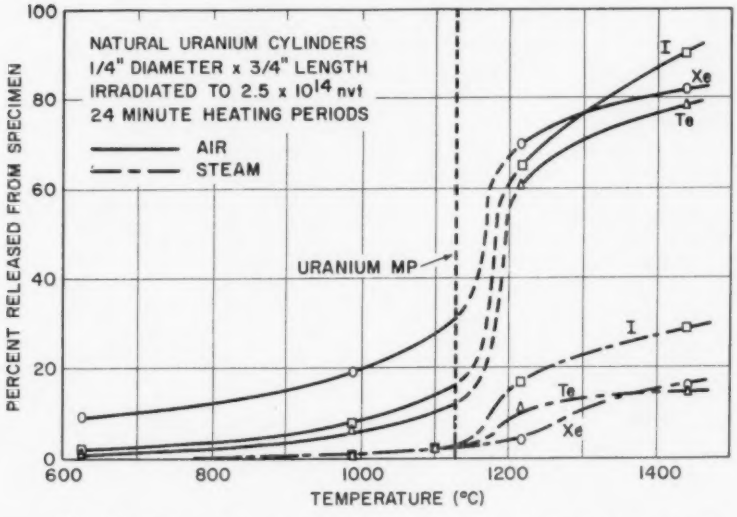


FIG. 3. Effect of temperature on release of volatile fission products.

was lower than in steam by factors of from $\frac{1}{2}$ to $\frac{1}{10}$.

One significant observation from Fig. 3 is that release of these elements is accelerated at temperatures above the melting point of uranium. The points of inflection on the curves are uncertain, but probably occur slightly above 1132°C, the melting point of uranium.

Effect of the oxidation reaction on release of fission products

The presence of an oxidizing atmosphere during heating of uranium influences the release of fission products in two ways. First, the fission products have opportunity to react with the oxidant and thus change their chemical composition and volatility. Tellurium, for example,

which boils at 1390°C in the elemental form, oxidizes readily to TeO_2 which sublimes at 450°C. Thus an oxidizing atmosphere should increase the release rate of tellurium. In air and steam atmospheres this was the observed case, with release rates comparable to those of iodine and xenon. In a helium atmosphere the release was less than 1 per cent. On the other hand, formation of a refractory oxide should reduce the volatilization rate. Strontium, for example, boils at 1380°C while the stable oxide SrO boils at a much higher temperature. Strontium release in air and steam was found to be somewhat lower than in helium.

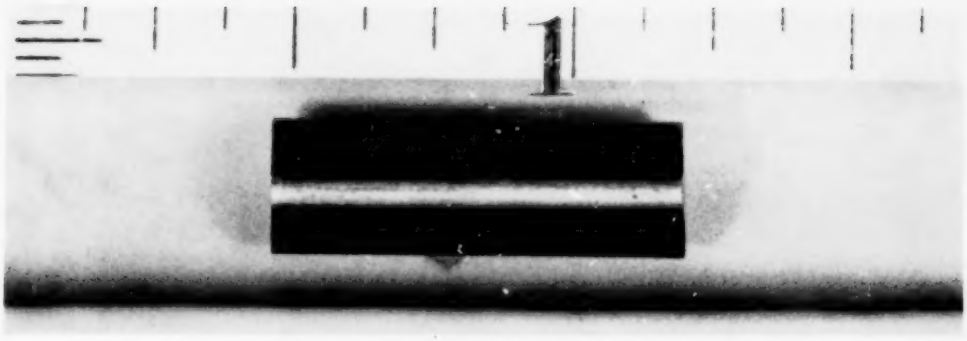
The second effect of an oxidizing atmosphere on the release of fission products relates to the changes that occur because of the oxidation of the uranium. Oxidation of uranium is quite rapid in air at high temperatures.[6] It is less in steam, but still appreciable.[8] The diffusion rate of fission-product nuclides to the oxide–metal interface would be independent of the type of gaseous atmosphere in which heating occurred. However, the radius of the metal cylinder is decreased constantly as the oxidation reaction progresses. Whether fission products would be released at higher or lower rates because of the oxidation would depend on the relative diffusivities in metal and oxide. Theoretical treatment of the diffusion problem is complicated by the fact that the physical properties of the oxide vary greatly depending upon the temperature and type of oxidizing atmosphere. One would hypothesize that if the oxide were porous, diffusion through the oxide would be more rapid than through the metal and therefore a higher release rate would result. Conversely, if diffusion through the oxide is slower than in the metal, the release rate would be reduced.

The oxide formed in air was primarily U_3O_8,[6] whereas that formed in steam was UO_2.[8] Though both types of oxide were sintered and adherent, that formed in steam appeared to be denser and more continuous than that formed in air. Photographs of specimens after oxidation in steam and in air at 1215°C are shown in Fig. 4. In these photographs, the air-treated specimen had been exposed for 24 min, resulting in 70 per cent oxidation. The steam-heated specimen had been exposed for 48 min, resulting in 18 per cent oxidation. The cracking and splitting shown in the photographs are believed to have occurred during cooling.

The experimental results indicate that diffusion through oxide formed by air is more rapid than through uranium and, in fact at 1215°C, the release rate is nearly proportional to the rate of oxidation. The same conclusion can be made for steam-produced oxide, but to a lesser degree. To illustrate this point, Fig. 5 was constructed, plotting the percentage release of volatile fission-product elements as a function of per cent of uranium oxidized. The experimental points are for temperatures above the melting point of uranium. At lower temperatures the correlation is less definite, though the release is still much greater than that calculated by diffusion from the metal. The present investigation dealt with uranium irradiated to very low levels. It is possible that higher burn-up would alter the diffusion rate in uranium and possibly increase both oxidation and release rates as functions of time and temperature.

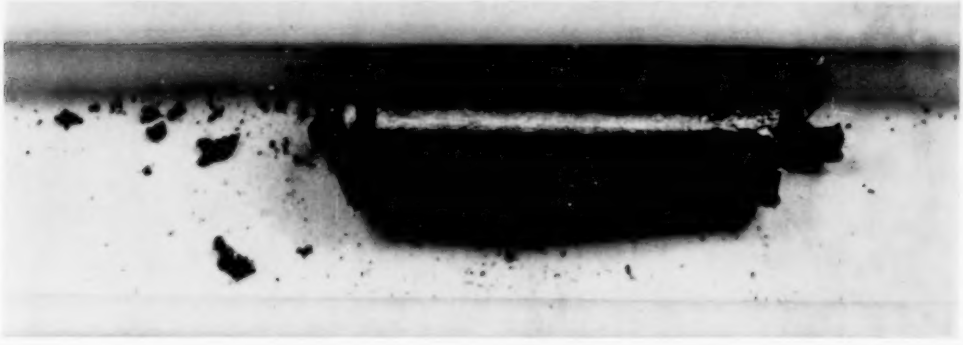
A further illustration of the influence of the oxidation reaction on release rates of the three volatile fission products is offered by Fig. 6. Here the oxidation of uranium in steam and in air is plotted as a function of time with temperature as a parameter. These data were collected by measuring the weight loss of uranium after removing the oxide by pickling in nitric acid. The higher oxidation rate in air is readily apparent. Comparison of this figure with Fig. 2, which gives the release of volatile elements with respect to time, shows a striking similarity.

An interesting aspect of the oxidation effect was observed during tests in a helium atmosphere. Oxidation by traces of air remaining in the combustion system and the helium feed amounted to 1–4 per cent of the uranium. The thin oxide film was found to build up in fission-product concentration rather than to be depleted as was the case with the metal. The enrichment of the oxide layer increased with time of heating to from ten to twenty times the original concentration. Concurrently, the average concentration in the metal decreased by diffusion to as low as one-tenth its original value.



(a) Irradiated specimen before heating.

(b) After heating in air at 1215°C for 24 min.



(c) After heating in steam at 1215°C for 48 min.

FIG. 4. Photographs of uranium test specimens.

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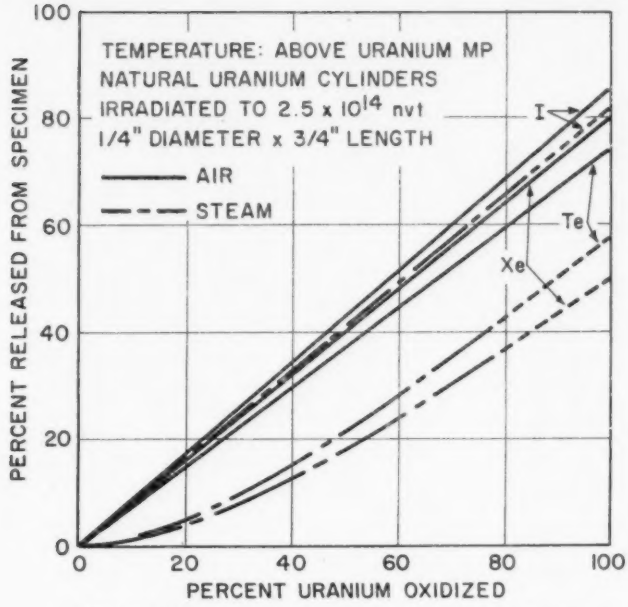

FIG. 5. Effect of uranium oxidation on release of volatile fission products.

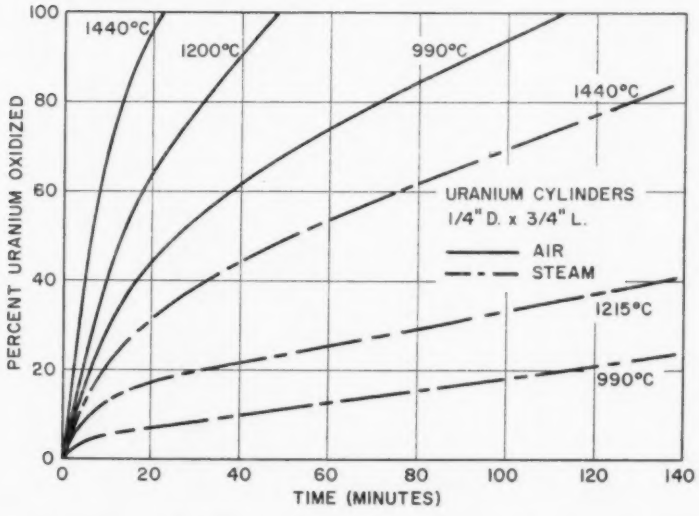

FIG. 6. Effect of time on uranium oxidation at various temperatures.

Table 2. Deposition of released fission products
(Uranium heated at 1215°C for various times)

Atmosphere	Average per cent of released material deposited in furnace tube						
	I^{131}	Te^{132}	Sr^{89-90}	$Cs^{136-137}$	Ru^{103}	Ba^{140}	Zr^{95}
Air	10	20	55	40	85	50	40
Steam	60	70	50	45	60	85	50
Helium	90	98	90	50	60	98	40

The release of iodine, tellurium and xenon in the helium tests were lower than was expected by diffusion calculations. Release from 0.2 to 7 per cent was observed. Entrapment in the oxide can be compared to high temperature pyroprocessing methods using oxide drossing techniques.

Behavior of released fission products

The foregoing discussions have dealt with the release of fission products from the uranium specimen. Fission products that are released from a fuel will not leave the reactor quantitatively but will be held within the reactor core, biological shield and associated ductwork by condensation and deposition. Some knowledge of the tendency for retention of released fission products to be deposited on tube walls was obtained from the laboratory experiments. The effluent gas stream swept the evolved fission products through a 15-in. section of the 1-in. inside diameter furnace tube. The wall of this tube section varied in temperature from slightly less than that of the heated specimen to about 50°C. The amount of each fission-product isotope remaining in this section of the furnace tube was determined, thus enabling a calculation to be made of the fraction of the volatilized material deposited therein. Table 2 lists the average results of all the tests made at 1215°C. At lower heating temperatures more deposition usually occurred. This table shows that a significant portion of the released material tends to deposit as soon as it leaves the heated area.

Reactor containment evaluation was not within the scope of these experiments; however, the experimental techniques lent themselves easily to qualitative evaluation of filter efficiencies for retaining evolved fission products. "Absolute" filters were located at the furnace tube outlet in several of the tests. They were MSA fiberglass ultra media supported by a Millipore filter holder, and rated at 99.95 per cent retention of 0.3-μ particles or larger. The amount of each isotope collected on the filter medium was measured separately. Table 3 lists the per cent of the released material reaching the filter which was retained on the filter medium. It shows that from 50 to 99 per cent of the gas-borne fission products were removed. No attempt was made to determine the physical

Table 3. Filter efficiencies for removal of fission product aerosols and vapors
(Uranium heated at 1215°C with "absolute" filter at furnace tube outlet)

Atmosphere	Per cent of released material retained by filter					
	I^{131}	Te^{132}	Sr^{89-90}	$Cs^{136-137}$	Ru^{103}	Ba^{140}
Air	56	97	53	93	97	73
Steam*	98	99	80	55	92	74
Helium	98	99	38	49	78	50

* Filter temperature 140°C.

properties of the collected material. Both condensed vapors and particulates were probably retained on the filter. In steam atmospheres, the filter was maintained at 140°C to assure that steam condensation did not occur on the filter surface.

In the tests with steam atmospheres, the off-gas steam was condensed after leaving the furnace tube. It was interesting to note that a very large percentage of the released fission products was contained in the condensate catch strongly dependent on extent of oxidation of the uranium.

Release of the volatile elements was greatest in air, next greatest in steam and least in helium atmospheres for similar conditions of time and temperature.

All fission-product elements except rare gases escaping the heated uranium tended to deposit heavily on tube walls. From 10 to 98 per cent of the released material deposited in the furnace tube used in the present study. When heating

Table 4. Comparative effectiveness of steam condensation vs. filter in removing released fission products

Method of removal	Per cent fission product removed					
	I^{131}	Te^{132}	Sr^{89-90}	$Cs^{136-137}$	Ru^{103}	Ba^{140}
By "Absolute" filter*	98	99	80	55	92	74
By steam condensation†	97	77	40	80	60	50

* Filter temperature 140°C.

† Average of six experiments, uranium heated at 1215°C in steam.

borne fission products by condensation of the fission product laden steam is nearly as effective in removing the fission products as an "absolute" filter.

Summarizing the behavior of the released fission products, it can be stated that for the conditions employed in these tests, deposition on cool tube walls, retention on filters and condensation of steam are effective mechanisms for retaining the evolved radioactive materials within the reactor shell or building.

SUMMARY

Release of strontium, barium and zirconium from heated uranium was lower than most previous theoretical estimates, being <0.5 per cent under all conditions tested.

Cesium and ruthenium behaved as "semi-volatile" elements being released from <1 per cent to about 15 per cent. Their release was dependent on time, temperature and atmosphere.

Xenon, iodine and tellurium were volatile, their release from heated uranium being dependent on time, temperature and atmosphere.

The release of the three volatile elements was flask. Table 4 shows that the retention of steam-occurred in steam atmospheres, condensing the steam caused 40–97 per cent of the evolved fission products to remain in the condensate.

"Absolute" filters removed 50–98 per cent of the evolved fission products, the efficiency depending on the isotope and the type of gas used.

The test parameters used in these experiments tend to maximize the release of fission products. One exception to this is the possibility that at higher irradiation levels, release could possibly be greater due to high concentrations of fission products and radiation damage to the uranium.

This laboratory investigation shows that definite trends and correlations are obtainable for application to the fission-product release question. However, the many complicating factors involved necessitate a conservative attitude when applying the data to hazards evaluations.

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SOME TRENDS AND RECENT HEALTH PHYSICS INSTRUMENTATION DEVELOPMENTS AT HANFORD*

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Abstract—The radiological instrument development work of the Hanford Laboratories is reviewed with emphasis on particular classes of instruments to demonstrate the present thinking and the trends. In general, two groups of instrumentation are considered: The first is survey and monitoring instruments and the second is special purpose instruments. Photographs and descriptions are included.

The survey and monitoring instruments include transistorized scintillation hand and shoe counters, an analyzing stack effluent radioisotope monitor, α-air monitors, transistorized scintillation portable survey instruments, and trends in environmental monitoring systems.

The special purpose instruments are those which are used in biology and medicine programs other than instrumentation. These include an all automatic data gathering and radio telemetering system for meteorological and radiological purposes, an instrument for rapidly determining the number of zinc sulfide particles on a filter for meteorological experimentation, a solar radiation integrator for automatically averaging the output signal from a pyrheliometer, and instruments for delineating the radioactivity in various types of experimental animals.

INTRODUCTION

THE purpose of this paper is to describe some of the philosophies and work of the Radiation Protection Instrument Research and Development Operation at the Hanford Atomic Products Operation, a General Electric-operated prime contractor for the United States Atomic Energy Commission. The Radiation Protection Instrument Research and Development Operation combines the efforts of the equivalent of approximately four men to investigate the techniques and concepts associated with health physics instrumentation and to demonstrate applicable techniques with experimental and prototype instruments. The work is reviewed with emphasis on particular instruments which are considered as representative and which might be of most interest to members of the Health Physics Society.

The two general classes of instruments described include survey and monitoring instruments and special purpose instruments.

SURVEY AND MONITORING INSTRUMENTS

The examples with survey and monitoring instruments include portable survey instruments, hand and shoe counters, α-air monitors, a stack effluent radioisotope monitor, and some remarks on trends in environmental monitoring.

Portable survey instruments have received considerable emphasis in recent years because of the need for higher reliability and ease of maintenance. The use of many different types of instruments for a given job has resulted in the need for a comparatively high spare-parts inventory; hence, a study of portable instrument standardization was started and is now coming to a close.[1] This study involved the investigations and application of scintillation detection and of transistor and printed circuit techniques. By utilizing relatively few different standard components in various combinations, instruments can be made for the detection and measurement of α, γ, β–γ, slow neutron, and fast neutron activity—and

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† Operated by General Electric Company for the United States Atomic Energy Commission.

for special purposes such as γ-energy analysis. Figs. 1 and 2 show some examples of experimental and prototype instruments with these components. Fig. 1(a) shows a transistor G–M instrument with a loudspeaker, meter and phone jack. Also included in Fig. 1(a) is a high-voltage supply and two plug-in printed circuit boards of the type used. Fig. 1(b) shows a scintillation transistorized α-poppy and count-rate meter. Headphones are used for very low-level, 100 dis/min or less, α-detection, and the meter can be read for the higher levels. The instrument in Fig. 1(c) combines the functions of the α and the β–γ meters. Figs. 1(d) and 2(a) are of slow and fast neutron detectors, respectively, Fig. 2(b) shows a portable γ-energy analyzer, and Fig. 2(c) shows a scintillation γ-dose-rate meter. The standard components utilized in these instruments include an amplifier, a count-rate meter circuit, a loudspeaker driver circuit, a high-voltage supply, and various types of scintillation detectors. The γ-energy analyzer uses also a window-width discriminator.

The work on hand and shoe counters illustrates applications of the investigations on scintillation techniques and on transistorization and standardization.[2] A scintillation transistorized α-hand counter was developed and an experimental model was put into actual use in field tests by engineers and radiation monitors for approximately 1 year. The instrument was found to be satisfactory and highly reliable. A scintillation transistorized combination instrument was then developed which can detect α-contamination on the hand surfaces and β- and γ-contamination on the hand and shoe surfaces. Fig. 3(a) shows the combination instrument between the two instruments which it can replace. The combination instrument is a table-top model with cable-connected shoe probes, and with an additional cable-connected hand-held probe for surveying clothing. The instrument on the left in Fig. 3(a) is the presently used fourfold α-counter and that on the right is the fivefold β–γ-counter. Fig. 3(b) shows a portion of one of the hand probes in the new counter. A side-window photomultiplier tube is cradled in a lucite light-pipe, the flat surface of which has a bioplastic scintillator layer for β- and γ-detection and a sprayed zinc sulfide coating for α-detection. The entire probe surface is useful for each type of radiation since the whole surface is covered in layers by each type of scintillator. The outer scintillator layer is covered by a thin layer of aluminium-coated mylar to provide a light shield.

Development effort has continued for several years on techniques associated with air α-contamination detection and monitoring systems.[3] The first instrument developed was a low air-flow high-level air α-monitor, a photograph of which is shown in Fig. 4(b). Air drawn through a fixed filter at the rate of 2 ft^3/min deposits particulate contamination which is viewed by a scintillation α-detector. The components of the detector can be seen in Fig. 4(a). A recent application of two of these instruments resulted in a sensitivity improvement by a factor of approximately 25. The sensitivity limit on the high-level monitor is imposed directly as a result of natural background radiation. Since the background effects is expected to be the same on two instruments located in the same general vicinity, the difference in their outputs eliminated much of the background effect. Fig. 5(a) shows a dual instrument which has two separate and independent high-level air α-monitors in addition to differential circuitry, recording, and alarm circuits. The high-level alarms are set at about 8000 dis/min and the differential alarm is set at about 300 dis/min. In this particular application, there was little probability of contamination being present at both detector locations at the same time, but the background effects are alike. The differential alarm will actuate in about 20 min for a contamination level of one hundred times the maximum permissible concentration. Another instrument with about the same sensitivity as the dual monitor, but using a single probe, is the medium-level air α-monitor shown in Fig. 5(b).[4] The alarm circuits on this instrument are actuated by fast rates of change of the filter activity, but are not actuated for slow rates of change. Work is in progress to eliminate further the natural background effects so that sensitivity will be determined almost entirely by air flow rates rather than by background.

An application of computer techniques to automatically provide data in a more usable form was demonstrated in the stack effluent radioisotope monitor, the block diagram of which is shown in Fig. 6(a).[5] Four signal outputs from scrubber and filter samplers are treated as four variables. The simultaneous analog solution of four equations with constant coefficients results in four outputs which are proportional to the quantities of four radioisotopes emitted out of the stack. While the present installation is used for only three variables, the concept of utilizing analog computing elements for such purposes is felt to be valuable. Analog, rather than digital counting, was found to be less expensive and less accurate. In the sample output record shown in Fig. 6(b), each pulse represents a specific amount of emission. The instrument also contains registers which continuously display the total number of pulses in each channel since the register was last reset.

The many improvements in detection and analyzing techniques in the last 5 years or so have opened the way for improvements in routine environmental

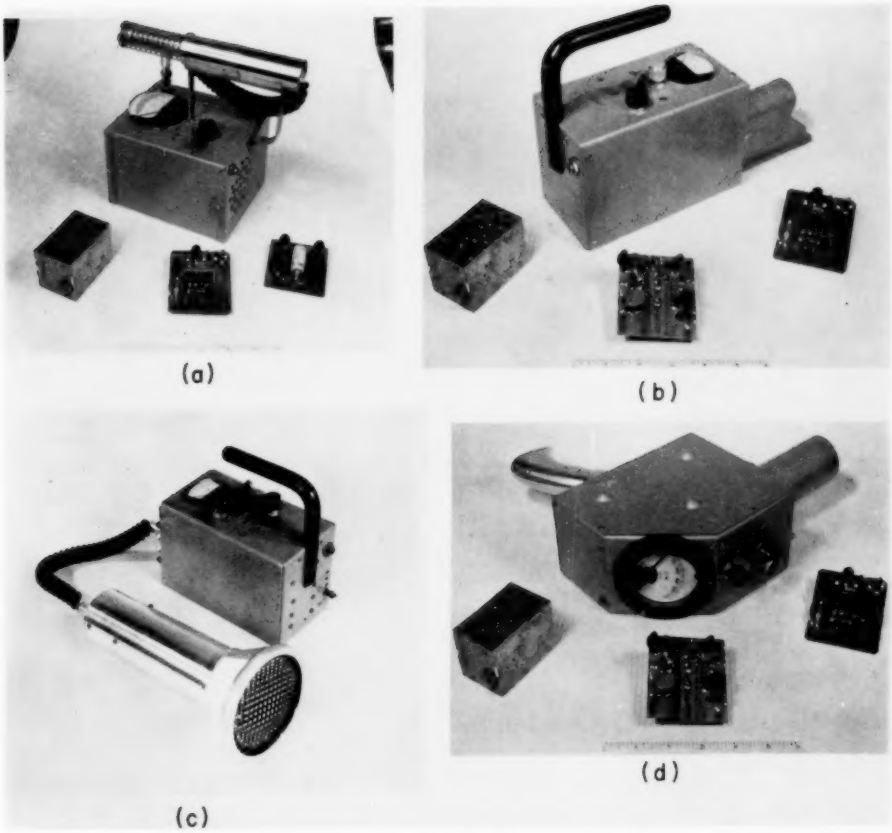


FIG. 1. (a) Loudspeaker G–M meter. (b) Scintillation α-meter. (c) Scintillation α-, β-, γ-meter. (d) Scintillation slow-neutron meter.

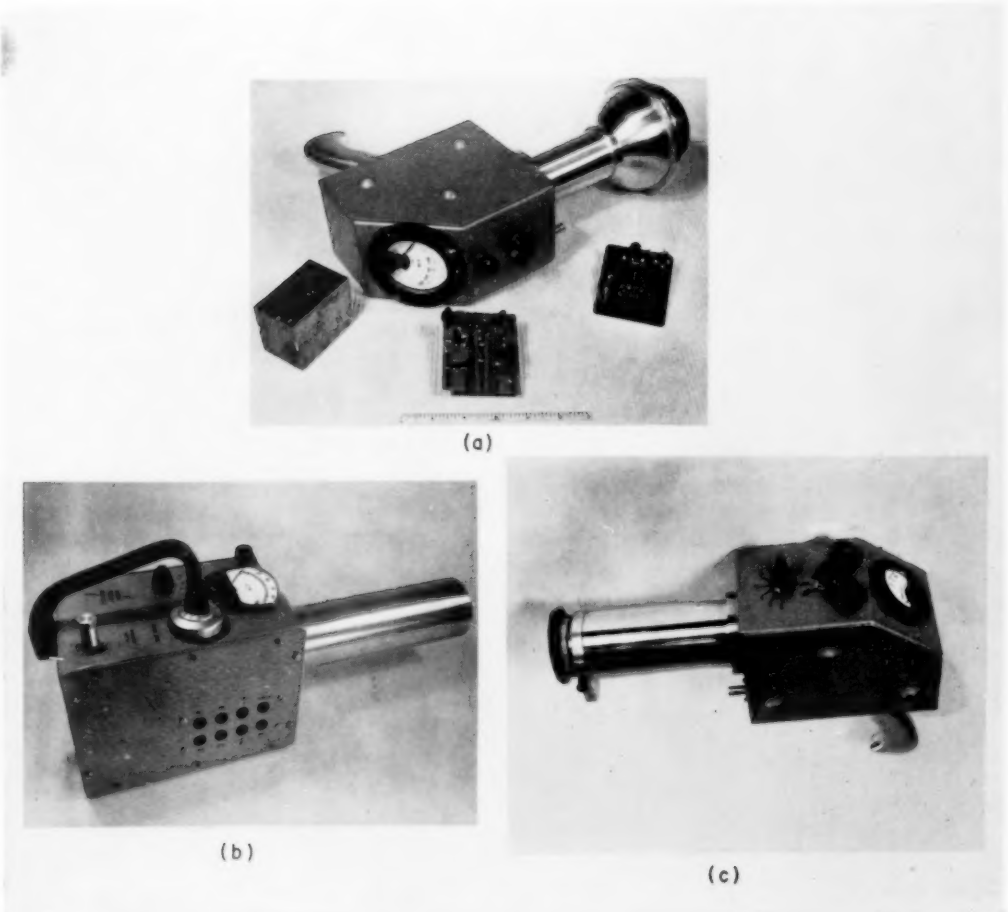


FIG. 2. (a) Scintillation fast neutron meter. (b) Portable γ-energy analyzer. (c) Scintillation γ-dose-rate meter.

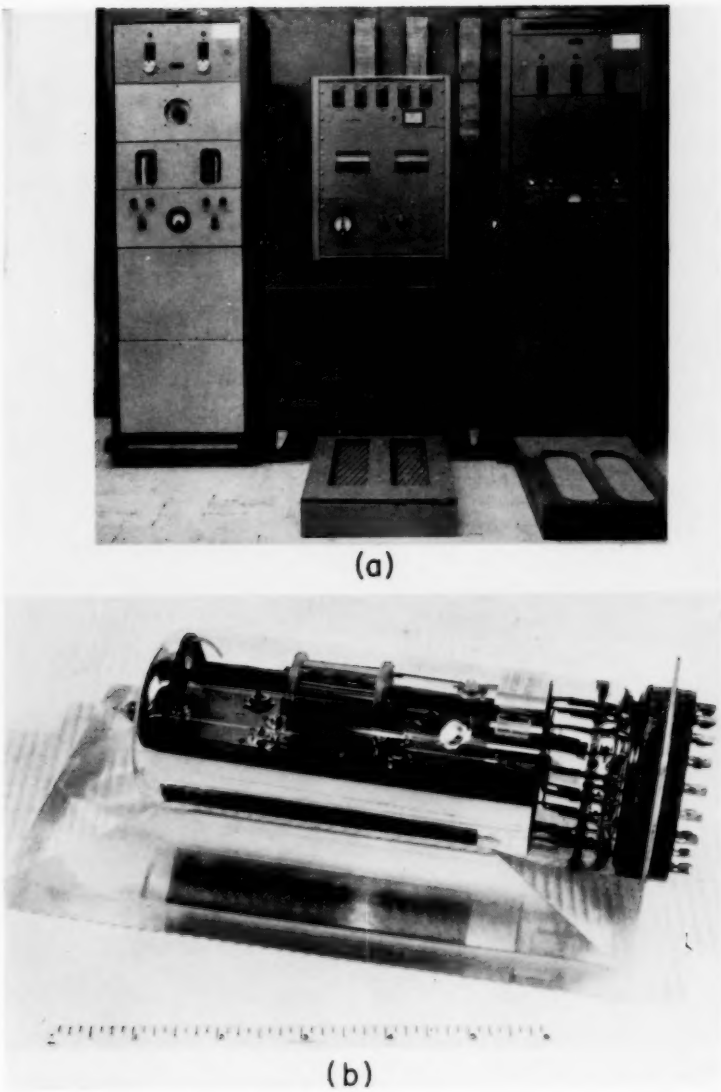


FIG. 3. (a) α-, β-, γ-hand and shoe counter (center instrument).
(b) Detector for scintillation hand counter.

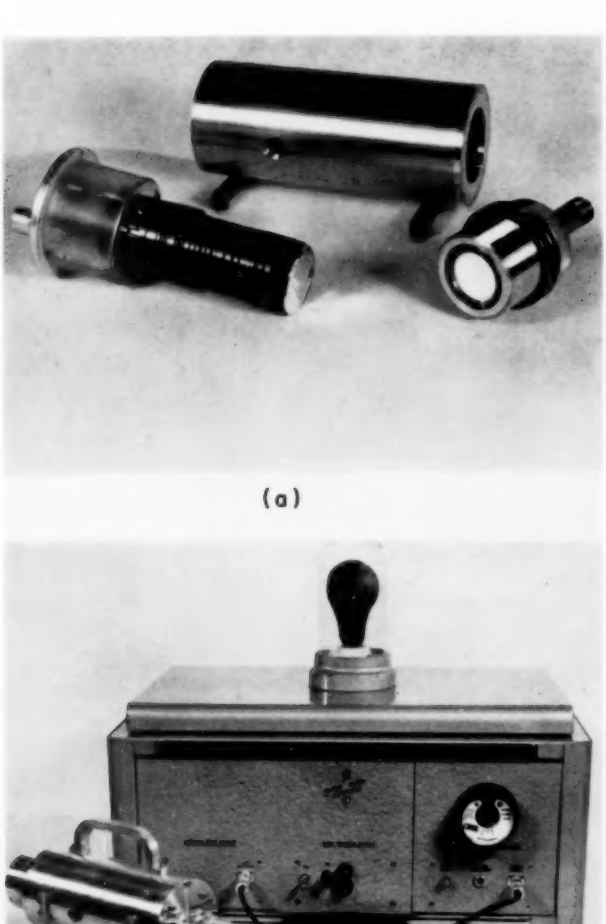


FIG. 4. (a) Air α-detector. (b) high level α-air monitor.

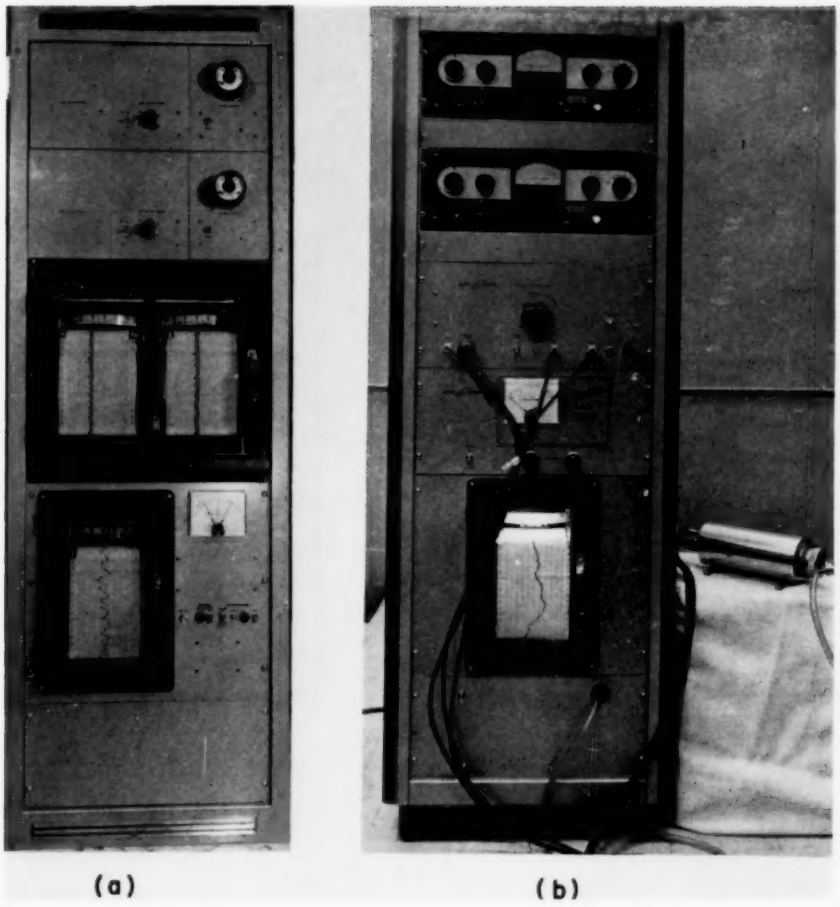


FIG. 5. (a) Dual air monitor. (b) Medium level air Alpha Monitor.

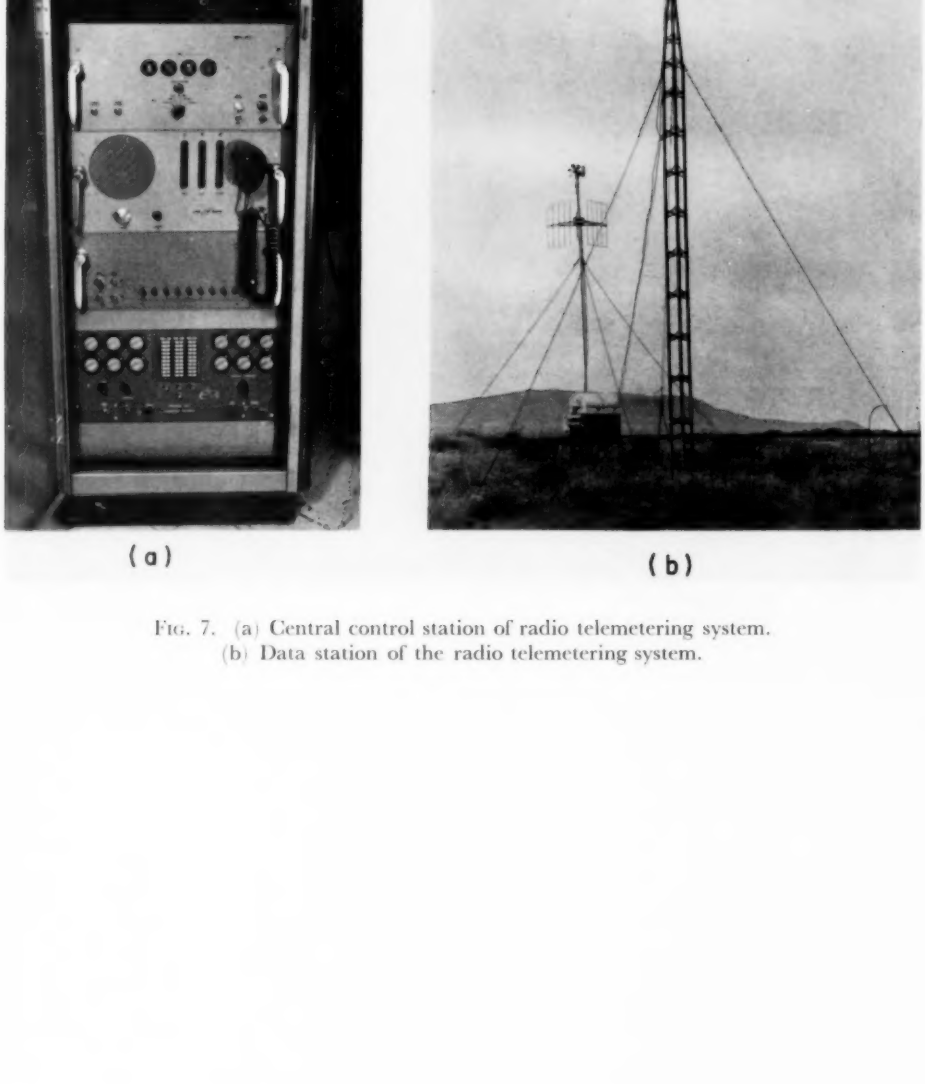


FIG. 7. (a) Central control station of radio telemetering system.
(b) Data station of the radio telemetering system.

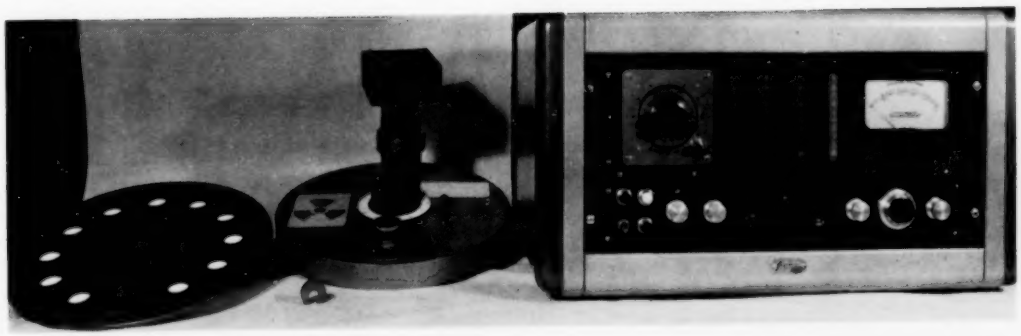
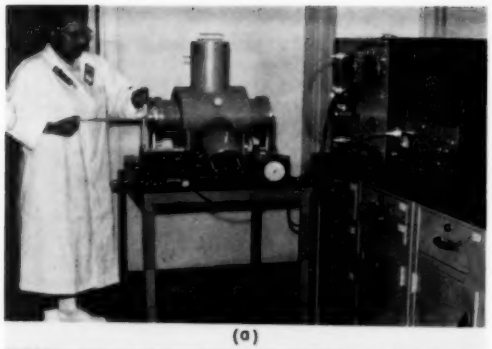
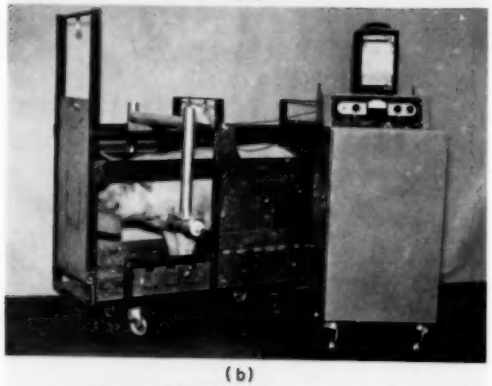


FIG. 8. Zinc sulfide particle counter.



(a)



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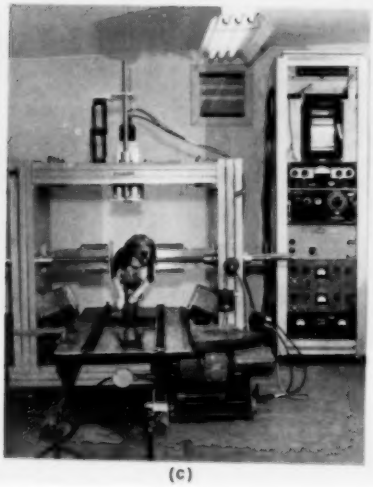


(c)

FIG. 10. (a) Mouse radiation counter. (b) Pig thyroid radiation counter. (c) Dog radiation counter.

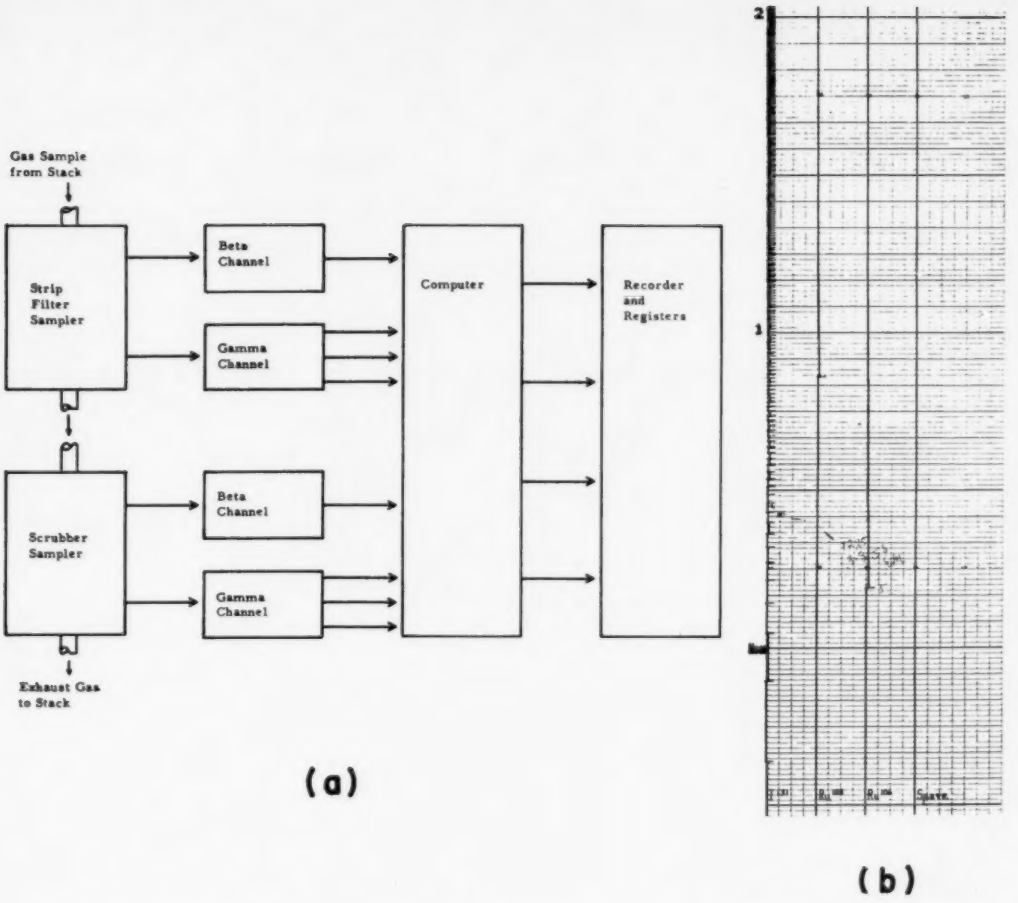
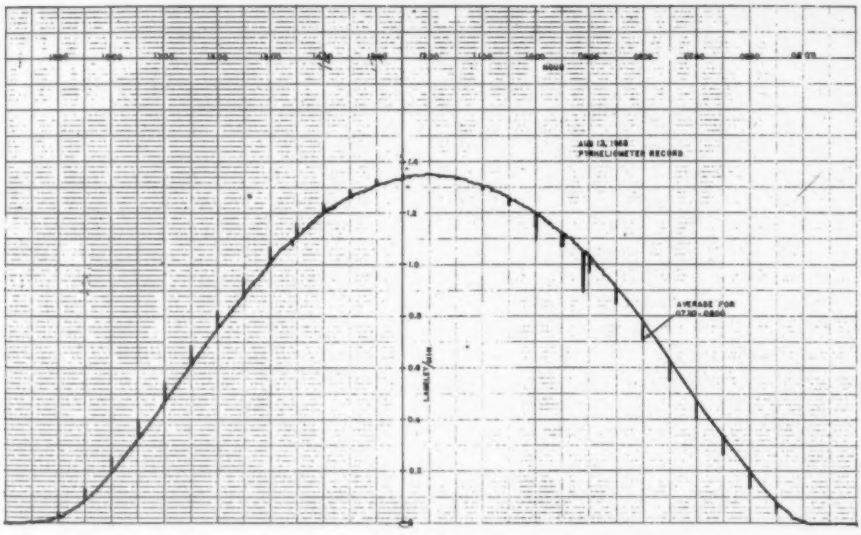


FIG. 6. (a) Block diagram of a stack effluent radioisotope monitor. (b) Output record showing an emission of I^{131}.

monitoring equipment. With a given reliability, we can detect lower amounts of some types of radiation now than ever before. Recent emphasis by the Atomic Energy Commission on improved continuous monitoring programs prompted us to look at the various techniques and devices available and to consider how they might best be utilized. The present trends at Hanford point to environmental and personnel monitoring systems which provide automatic and continuous communication of background radiation conditions and of dose rates and total doses received by personnel working in the vicinity. Alarms for non-standard conditions are expected to be included. Our experience with detectors and the associated circuitry allow us to provide designs which are generally better than our present continuous monitoring instruments; hence, development effort is expected to consider techniques and concepts related to communication channels and the methods of presentation.

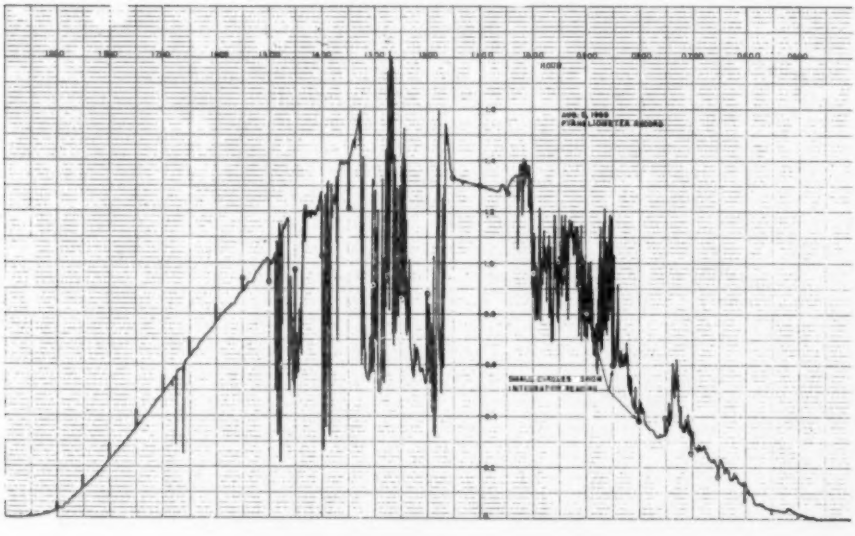
SPECIAL-PURPOSE INSTRUMENTS

The special-purpose instruments are those which are used in biology and medicine programs other than instrumentation. These include an automatic data-gathering and radio telemetering system for meteorological and



(a)

FIG. 9. (a) Pyrheliometer record for a clear day.



(b)

(b) Pyrheliometer record for a partly cloudy day.

radiological purposes, an instrument for rapidly determining the number of zinc sulfide particles on a filter for meteorological experimentation, a solar radiation integrator for automatically averaging the output signal from a pyrheliometer, and instruments for delineating the radioactivity in various types of experimental animals.

The fabrication and installation of the components for the radio tetemetering system is nearing completion. The system has twenty data-gathering stations, located in and near the Hanford project, which are automatically interrogated once each hour by a radio signal from the central control station shown in Fig. 7(a). Each data station, Fig, 7(b), sends back information on the average values of the wind velocity vector components and the radiation levels since the last reporting time. The interrogation frequency can be changed to once every 30 min, 15 min, or 5 min by a switch on the central control station. The information is automatically typed out by a flexowriter for immediate reading and is punched out on paper tape for later analysis on an IBM digital computer.

The zinc sulfide particle counter is an instrument which was developed for use in the experimental meteorology program to automatically count the number of zinc sulfide particles on a small filter.[6] The particles are activated by a fixed-strength source of α-radiation and the resulting scintillations are counted with a photomultiplier and the associated amplifying and counting equipment. The observed counting rate is proportional to the number of particles on the filter. Fig. 8 shows a photograph of this instrument.

A solar radiation integrator was developed for use in the routine determination of average values of solar radiation incident upon the detector.[7] A recording of the detector output on a clear day is shown in Fig. 9(a) and that on a partly cloudy day is shown in Fig. 9(b). The previous method of obtaining hourly averages was to estimate by eye from the chart. A low-pass RC filter with a 15-min time constant is utilized for smoothing and the filter output is automatically printed on the chart during a 1-min interval every $\frac{1}{2}$ hr. This has resulted in a considerable time reduction for this operation on partly cloudy days.

The work on a mouse radiation counter, Fig. 10(a), was originally completed in 1955. The mouse can be moved past an adjustable width slit formed by lead collimators to expose only a small portion of the animal to the three scintillation γ-detectors. The width of the slit can be adjusted from about 1 cm to a value large enough that the entire mouse is exposed to the detectors. Recent additions to the instrument allow the counting of plutonium in mice.

An instrument for measuring the amount of I^{131} in the thyroids of pigs is shown in Fig. 10(b).[8] The instrument consists of two scintillation γ-detectors which supply signal pulses to an amplifier, a window discriminator, a counting-rate meter, and a recorder. This instrument has been in use since 1956.

Fig. 10(c) shows a radiation counter for determining the location and the extent of radioactivity in experimental dogs and other animals of about the same size. The two detectors shown near the dog are for detecting Pu^{239} or Sr^{90} and the other three detectors, not being used in this photograph, are for detecting γ-emitting isotopes such as Ru^{103}, Ru^{106} and Cs^{137}. The associated instrumentation consists of commercially available amplifiers, pulse-height analyzers, count-rate meters, recorders and power supplies.

An experimental fish radiation counter was set up in the laboratory to determine the feasibility of measuring the amount and location of plutonium in live fish without killing them. In order to reduce the amount of absorbing material between the fish and the detector, the fish was held out of water and water was inserted into the mouth and allowed to flow out of the gills. The technique was found feasible and it is expected that an instrument will be fabricated soon for this purpose.

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STATUS OF STATE LEGISLATION RELATED TO IONIZING RADIATION

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Abstract—State legislation as it pertains to ionizing radiation has been studied. More than half of the states in the country have Radiation Protection Acts; many states are in the process of promulgating rules and regulations under the Acts. The educational process will be helpful in promoting an effective code, and training of qualified personnel to effectively carry out the laws will be needed.

A STUDY has been made of state legislation as it relates to ionizing radiation. In April 1960, requests were mailed to each of the fifty states inquiring about such legislation. Replies were received from forty-six states; thirty-three of which enacted some type of legislation. Some states have only industrial codes which do not apply to the licensed art of healing; for example, hospitals, clinics and physicians. Four states exempt those installations found to be without hazard as determined by standards set up by the National Committee on Radiation Protection, whereas another state exempts installations where radioactive materials are under control. In another state private offices employing radiation are exempt. One state, although not having enacted a Radiation Protection Act, has an active radiological health program conducted under authority of the Sanitary Code.

As of April 1960, twenty-one states have Radiation Protection Codes,* while sixteen states have Enabling Acts. Of the sixteen states with Enabling Acts, twelve are in the process of developing rules and regulations promulgated under the authority of their respective Enabling Acts. Further information regarding activities of individual states is presented in Table 1, which has been compiled from the Enabling Acts, Codes, and other pertinent information received from the states. In some instances where there is a Radiation Protection Code, it is not known whether there had been previous enabling legislation. Eleven states utilize the recommendations of the National Committee on Radiation Protection as standards. Thirteen states have Advisory Committees composed of scientific and medical personnel (such persons as radiologists, dentists, radiological physicists, and so forth), who serve as technical advisors to the Administrative Agency or the Enforcement Agency (usually the Board of Health) or both. The function of this committee varies with each state, but, in general, its work entails the reviewing of policies and the program relating to radiation matters, such as standards of practice and radiation instrumentation.

Provision is made in a large number of codes for the registration of radiation sources with the Administrative Agency. Radiation surveys and monitoring performed by the owners are required of such users who may hire a radiological physicist to carry out these requirements or have a qualified person within the organization conduct these surveys. The Administrative Agency in some states may review plans and specifications on the design and shielding for radiation sources. The Administrative Agency may make inspections of the radiation installations for the purpose of determining any possible radiation hazard. Under their codes a large

* Since the replies to the April 1960 questionnaire were received, additional information has been received from the following states: New Jersey and New Mexico have developed Radiation Protection Codes. Massachusetts has passed an Enabling Act.

Table 1. *Scope of legislation pertaining to ionizing radiation in the fifty states*

State	Legislation	Code	Authority	Registration	Duties			Comments
					Inspection	Education	Review plans and specifications	
Arizona	No reply							
Alaska	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
Alabama	None							
Arkansas	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
California		Yes	Division of Industrial Safety Department of Health					Will have registrations in the near future
Colorado	No reply							
Connecticut		Yes	Department of Health	Yes				
Delaware		Yes	Department of Health					Excludes medicine, dentistry, veterinary medicine where radioactive materials are under competent control
Florida		Industrial	Division of Industrial Hygiene		Yes			No specific legislation but an active radiological health program under authority of sanitary code
Georgia		Yes	Department of Health					Georgia Water Quality Control Act applies in part to disposal of radioactive materials Hospital Service Section of Health Department controls ionizing radiation in hospitals and nursing homes
Hawaii	No reply							
Idaho	None							
Illinois	Enabling Act	Industrial	Department of Health Department of Labor	Yes	Yes	Yes	Yes	
Indiana	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
Iowa	No reply							
Kansas	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
Kentucky		Yes	Department of Health	Yes				
Louisiana	No reply							
Maine		Yes	Department of Health	Yes	Yes			
Maryland	Enabling Act		Department of Health					Division of Occupational Health has been engaged in control of ionizing Radiation in the past decade
Massachusetts		Yes	Department of Labor and Industries	Yes	Yes			Code does not apply to hospitals, sanitoria, medical clinics, dental clinics and veterinary clinics
Michigan		Yes	Department of Health	Yes				
Minnesota	Enabling Act	Yes	Department of Health	Yes				
Mississippi		Yes	Department of Health	Yes				Narrow application, applies only to industry
Missouri	None							
Montana		Yes	Division of Industrial Hygiene					Inspection of non-AEC controlled radiation sources
Nebraska	None							
Nevada	None							
New Hampshire	None							
New Jersey	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
New Mexico	Enabling Act		Department of Health	Yes				
New York		Yes	Department of Health	Yes				No jurisdiction over New York City, which is covered by Code of New York City Dept. of Health
North Carolina	Enabling Act	Industrial	Department of Labor Department of Health	Yes Yes	Yes	Yes		
North Dakota		Yes	Department of Health					By-product material licensed by AEC excluded
Ohio	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	
Oklahoma	Enabling Act		Department of Health	Yes	Yes	Yes	Yes	Excludes healing arts, veterinary medicine, or duly licensed practitioners
Oregon	Enabling Act		Department of Health					
Pennsylvania		Yes	Department of Health	Yes				
Rhode Island	None							

Table 1—(*contd.*)

State	Legislation	Code	Authority	Registration	Duties			Comments
					Inspection	Education	Review plans and specifications	
South Carolina		Yes	Department of Health	Yes	Yes			Cope applies only to Fluoroscopic Shoe Fitting machines
South Dakota	Enabling Act	Yes	Department of Health	Yes	Yes	Yes	Yes	
Tennessee	Enabling Act		Department of Health	Yes	Yes	Yes		
Texas		Yes	Department of Health	Yes				
Utah			Department of Health	Yes				Registration program for radioisotopes
Vermont	None							
Virginia			Department of Health	Yes				Act for registration of sources not controlled by AEC
Washington	Enabling Act		Many					Nuclear Energy Legislation
West Virginia	None							
Wisconsin		Yes	Industrial Commission					
Wyoming	None		Department of Health					Registration of users of isotopes and producers of source material

number of states require an accumulation of information on radiation protection and control and its subsequent dissemination within the state. This last provision is extremely important in having an effective radiation code that allows much progress to be made by education.

Within the next 2 years most of the state agencies will have promulgated their rules and regulations. Trained personnel will be needed to carry out the programs. These persons must be trained in all of the radiation concepts relating to protection; they must be familiar with radiation instrumentation; they must be familiar with the varied techniques in the field; they must have enough experience to evaluate each situation and make proper interpretations of the data; and they must be able to make suitable recommendations.

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TRICHLOROETHYLENE-SATURATED WATER AND TETRACHLOROETHYLENE CHEMICAL DOSIMETER SYSTEMS—NEUTRON RESPONSE*†

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Abstract—The response of the trichloroethylene-saturated water system has been found to be dependent on neutron energy in the region from 1 to 14 MeV. However, the response to fission neutrons with spectra of different degrees of degradation appears relatively constant. Measurements of neutron dose with this type of dosimeter can be made, subject to the limitations of the subtractive method, the experimental error, and calibration for neutron sensitivity. The low sensitivity to fast neutrons of the tetrachloroethylene dosimeter makes it valuable for the measurement of γ-dose in a mixed radiation field. Both types require suitable shielding from thermal neutrons.

INTRODUCTION

TISSUE dosimeters are needed which are capable of measuring the individual contributions of each component of a mixed radiation field to the total dose. Such dosimeters should have a tissue-like energy dependence and, in addition, should be independent of dose rate. For biological systems, they should be capable of measuring the dose delivered to tissue and, for certain purposes (such as depth-dose determinations), be physically small enough that the dosimeter itself does not perturb the dose to the system being studied.

Chemical dosimeters, as developed thus far, appear to offer a distinct potential for fulfilling some of these requirements. The types of chemical dosimeters chosen for investigation have been discussed;[1] the preparation (essentially according to methods of SIGOLOFF[2]), response to γ-radiation, evaluation and other properties of the dosimeters used in this study have been reported earlier.[3] The systems appear generally applicable for measurement of γ-dose in the range of approximately 10 to 10^6 rads. Various sources of neutrons have been used to determine the response of these dosimeters in order to evaluate their usefulness in mixed radiation fields.

METHODS

The chemical dosimeters

Two types of dosimeters have been involved (Fig. 1), one which responds to both γ-radiation and to fast neutrons, and another which responds to γ-radiation with only a slight response to fast neutrons. Chemical chain reactions induced by radiation liberate acid in both types, and the amount of acid formed is directly proportional to dose. The response may be evaluated by titration of the acid produced, by change in conductivity of the solution, or by change in pH measured electrometrically or colorimetrically. In these studies, a colorimetric pH indicator was always present, usually chlorphenol red (CPR).

The dosimeters which respond to both fast neutrons and γ-rays consist of trichloroethylene-saturated water containing the pH indicator; this one-phase solution is essentially all water. The dosimeters which are sensitive to γ-radiation and only slightly sensitive to fast neutrons consist of tetrachloroethylene, which contains a small amount of free-radical chain-reaction retarder, overlayered with an aqueous phase containing the indicator. The sensitivity of both types depends on the concentration of chain-reaction retarders and, with spectrophotometric evaluation, on

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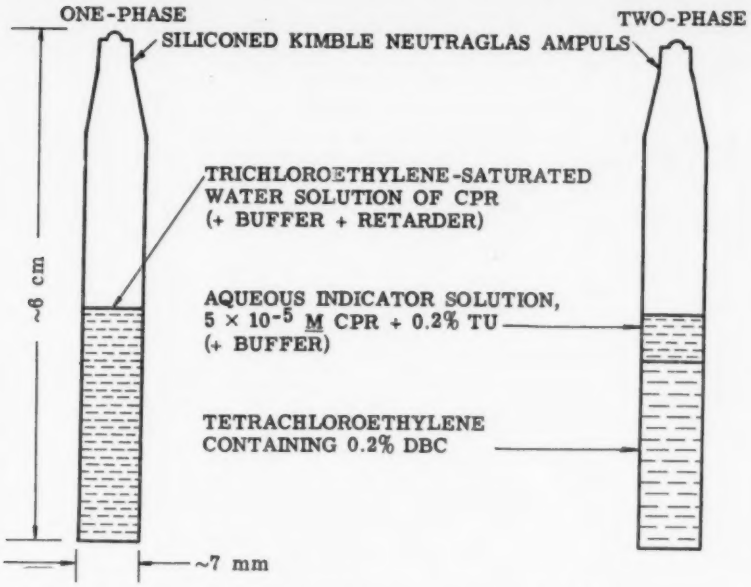

FIG. 1. One-phase and two-phase chemical dosimeters.

the presence of buffers and the initial pH of the aqueous phase. In the one-phase type thiourea (TU) is used as a retarder, and for the two-phase dosimeters, 2,6-di-tert-butyl-*p*-cresol (DBC). The solutions (0.75 ml) are contained in 1-ml flame-sealed ampuls.

Evaluation

The most precise and convenient method for determination of response (i.e. the amount of acid produced by radiation) is by spectrophotometry. The indicator has two widely separated absorption bands in the visible region of the spectrum. The acid form of chlorphenol red absorbs at 433 mμ (producing a yellow color), and its conjugate base absorbs at 570 mμ (giving a purple color). Over a certain pH range, both bands are present resulting in intermediate colors. The function which is used for evaluating the dosimeters is the ratio of the transmittance of the basic band to the transmittance of the acid band. The measurement of dose is made by considering the difference between the pre-exposure and post-exposure ratios. A simple two-band filter photometer has been found to be quite satisfactory for this purpose.[3]

The one-phase system, owing to its high hydrogen content, has a considerable response to fast neutrons in addition to being a good γ-dosimeter. By using the two-phase dosimeter with its very low response to neutrons, one should be able to use the paired system for the simultaneous determination of neutron and γ-dosage. The parameters involved in such a determination may be expressed by the following equations:

$$R_1 = D_\gamma + N_1 D_n = \text{response of one-phase dosimeter } (\gamma\text{-equivalent r})$$

$$R_2 = D_\gamma + N_2 D_n = \text{response of two-phase dosimeter } (\gamma\text{-equivalent r})$$

where $D_\gamma = R_2 - N_2 D_n = \gamma$-dose (r)

$$D_n = (R_1 - R_2)/(N_1 - N_2) = \text{neutron dose (rads)}$$

N_1 and N_2 are the neutron sensitivities of the one-phase and two-phase dosimeters, respectively, in terms of the number of roentgens of γ-radiation required to produce the same response as 1 rad of neutrons. These two constants must be known, in addition to the experimentally determined values of the responses of the two types of dosimeters for the particular measurement. In order to determine these constants, exposures to known doses of radiation were made.

EXPOSURES

Thermal neutrons

Both the one-phase and two-phase systems are sensitive to thermal neutrons. This was determined by exposures to known thermal neutron

and γ-doses in the Los Alamos Water Boiler.[4] It was found that 1 rad of thermal neutrons (2×10^{10} n/cm^2) produces a response with the one-phase system equivalent to 3.5 r of γ-radiation and to 4.5 r with the two-phase system. The effects of thermal neutrons are eliminated by shielding the dosimeters with $\frac{1}{2}$ in. of lithium metal contained in double-walled aluminum cans. Much smaller shields for individual dosimeters can be constructed using Li^6.

Monoenergetic neutrons

The response of both types of dosimeters to neutrons of various energies has been determined by exposure to known doses of neutrons of 1, 2, 4, 6, 8, and 14 MeV. The 14-MeV neutrons were obtained from the Cockcroft–Walton, and the other energies from the Van de Graaff machine. The $T(p,n)He^3$ reaction was used for 1, 2, and 4 MeV, and the $D(d,n)He^3$ for 6 and 8 MeV. The assumption had to be made that the γ-contamination was negligible; this is thought to be quite valid for both accelerators. For the Van de Graaff exposures, the dosimeters were attached directly to the back side of a U^{238} fission counter, which was used to measure the flux. In the case of the Cockcroft–Walton exposures, the flux was monitored by associated-particle (α) detection. The integrated dose was obtained from the flux measurements, exposure times, first collision theory, and appropriate geometry corrections. The results are given in Fig. 2.

It is obvious from these curves that energy dependence toward fast neutrons exists for these dosimeters. With neutron sources of known energy, suitable calibrations (such as these) could be used so that the dosimeters would be useful in dose evaluations.

Fission neutrons

The spectrum of fission neutrons from a reactor depends on its construction, amount of moderator, etc. Although the dosimeters showed energy dependence in the studies with monoenergetic neutrons, it was thought possible that they might not be greatly dependent on changes produced by various degrees of degradation in the broad spectrum of fission neutrons.

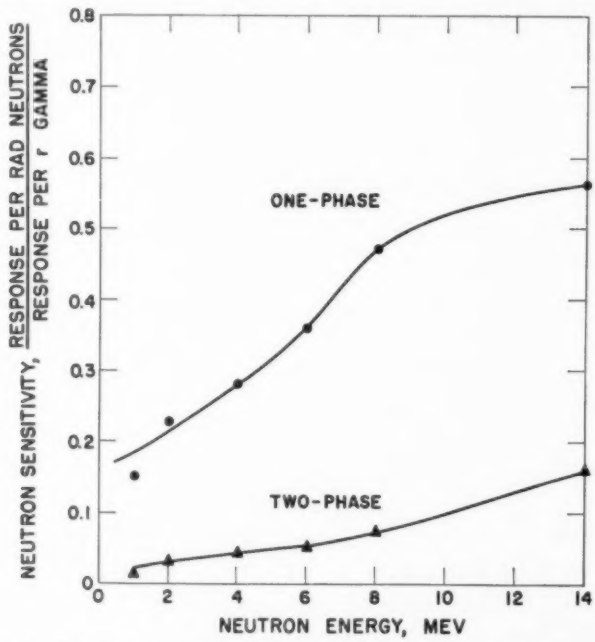

FIG. 2. Relative responses of chemical dosimeters to monoenergetic neutrons.

Exposures were made at the critical assemblies Godiva II[5] and Little Eva.[6] The Godiva spectrum was further degraded by shielding the dosimeters with various thicknesses of polyethylene. The tissue dose vs. energy distributions, as measured by the threshold detector techniques[5] utilizing Pu^{239}, Np^{237}, U^{238}, S^{32}, and first collision theory, are shown in Table 1.

Table 1. *Tissue dose spectra*

Energy interval (MeV)	Dose in energy interval (%)				
	Little Eva	Godiva $(CH_2)_n$ thickness (in.)			
		0	1	2	4
0.004–0.75	42	21	29	27	22
0.75–1.5	28	26	24	23	20
1.5–2.5	15	22	29	30	24
>2.5	15	31	19	21	34

It is likely with the greater thicknesses of polyethylene that a portion of the spectrum was degraded below the plutonium threshold, thus giving percentage dose values in the various energy intervals which are not indicative of the true spectrum.

Determination of the γ-component was also necessary. For Godiva, the γ-dose in free air was known to be approximately 10 per cent of the neutron dose[5] but was also observed to vary considerably, depending on the position of the assembly in the room and on the presence of other scattering materials. The method used to provide the value taken as the correct γ-dose for evaluation of N_2 employed a new type of liquid scintillation dose rate meter,[7] which is believed to have negligible response to fast neutrons. The two-phase chemical dosimeter was found to have a 4 per cent response to fast neutrons from Godiva (i.e. 100 rads of neutrons produced the same response as 4 r of γ-radiation). This number, although apparently small, is quite important at high neutron-to-γ dose ratios. The value of 0.04 for N_2 has been assumed to hold for all sources of fission neutrons and is used in subsequent calculations of γ-dose from response of two-phase dosimeters.

DISCUSSION

The results in Table 2 are indicative of the spread in values generally obtained using three dosimeters per measurement. It is possible, by exercising greater care and using more dosimeters per point, to make the determinations more precise. Owing to the limitations of the subtractive method, measurement of neutron dose is dependent on the accuracy of the γ-determination, as well as on the γ-to-neutron

Table 2. *Response of chemical dosimeters to degraded fission neutrons**

Polyethylene (in.)	D_n†	R_1‡	R_2§	$D\gamma$**	$R_1 - D\gamma$††	N_1‡‡
Godiva II						
0	1100	430	156	112	318	0.29
1	880	312	86	51	261	0.30
2	530	278	127	106	172	0.32
4	230	210	130	121	89	0.39
Little Eva						
0	660	286	34	8	278	0.42
0	380	173	26	11	162	0.43
0	230	102	18	9	93	0.40
0	170	66	10	3	63	0.37

* Dosimeters shielded by lithium.
† Fast neutron tissue dose (first collision rads in tissue).
‡ One-phase dosimeter response (γ-equivalent r).
§ Two-phase dosimeter response (γ-equivalent r).
** Gamma-dose $= R_2 - 0.04\,D_n$.
†† Response of one-phase dosimeters to D_n.
‡‡ Neutron sensitivity of one-phase dosimeter.

dose ratio. For example, when the γ-dose is ten times the neutron dose, the neutron response of the one-phase dosimeter is masked by the γ-response and within the experimental error.

The constant N_1 (neutron sensitivity of the one-phase dosimeter) has been observed to vary somewhat for a given type of dosimeter, prepared at different times but supposedly in the same manner. This difference probably arises from variation in concentration of some trace component, such as oxygen. The dose range can be extended by adding either buffer or retarder, or both. It appears that the chemical reactions produced by γ-radiation probably follow a different course than those initiated by neutrons, since cursory experiments at Godiva indicate that the neutron sensitivity is affected differently for range extension by buffer than by thiourea. The retarder seems to give increased neutron sensitivity, whereas buffer appears to decrease the response. These variations in neutron sensitivity, although not sufficiently investigated, do indicate that each preparation of one-phase dosimeters should be calibrated for neutron response; a γ-calibration and an assumed neutron sensitivity are not sufficient.

Acknowledgments—We are indebted to the critical assembly, large Van de Graaff, and Cockcroft-Walton groups for making the neutron exposures possible.

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RESPONSES OF SILVER-ACTIVATED PHOSPHATE GLASS TO α-, β- AND γ-RAYS AND NEUTRONS

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Abstract—The responses of Ag-activated phosphate glass to α, β, γ and neutron radiations have been examined. In each case there is a linear relationship between the glass reading and dose of ionizing radiation. The reading per rad in the glass is independent of photon or β-particle energy over a wide range, while at high LET it decreases with increasing LET, the reading being one-fifth as much per rad for α-particles as for γ-rays. The glass has been applied successfully in absolute measurement of absorbed dose in a β-radioactive solution, separate measurements of γ-ray dose and thermal neutron flux in a thermal column, and measurements of X-ray dose distribution in a mouse.

I. INTRODUCTION

A TOTAL radiation dosimeter approaches the ideal device if it is small and rugged; has linear response to the dose of γ-rays, α-particles, β-particles, neutrons or protons; has high sensitivity; has accurate reproducibility; and has negligible dependence on time, temperature and dose rate. This paper will show that silver-activated phosphate glass dosimeters have such properties.

SCHULMAN *et al.*[1] found that this glass exposed to X- or γ-rays acquires the ability to fluoresce when excited by ultraviolet rays and that the amount of fluorescence is linearly proportional to the exposure dose. He observed no fading effects at room temperature during 6 days after irradiation. Since then many studies[2–9] have been performed on the application of this glass to dosimetry. It has been proved that glass dosimeters can measure from 10^2 to 10^4 r for Co^{60} γ-rays with an error of less than ± 5 per cent and that their response per roentgen to low-energy X-rays depends sensitively on the X-ray energy. This energy dependence seems to be attributable to the energy dependence of the ratio of the X- or γ-ray mass absorption coefficients of the glass to the absorption coefficient of air to which the roentgen unit is referred.[9] Thus the glass response per rad in glass is almost independent of the energy of incident photons or β-particles except for very high or low energy.

This paper surveys the responses of the glass to X- and γ-rays, β- and α-particles, neutrons and recoil protons. For comparative studies among these responses, the term "response per rad in glass" or its equivalent is used or implied. Our approach is motivated more by a basic interest in solid state phenomena than in applied dosimetry where one would present the results in terms of the glass response vs. rads in tissue. The dependence, if any, of the response per glass-rad on the kind of radiation is attributable to the difference in LET (linear energy transfer) of the radiations. The relative response per glass-rad vs. LET presented in this paper indicates that the sensitivity of the glass continuously decreases with increasing LET in contrast to the RBE (relative biological effectiveness) vs. LET curve for chromosome aberrations (e.g. Ref. 10) and most other biological effects.

* On leave of absence from National Institute of Genetics, Misima, Japan, from Feb. 1958 to March 1959 as visiting atomic energy researcher sent by the Japanese Government.

† Operated by Union Carbide Corporation for the United States Atomic Energy Commission.

Small glass rods are suitable for implanting in materials where internal dose distributions are to be measured. Although the glass response per roentgen depends sensitively on the incident photon energy, the ratio of glass reading to ion chamber reading shows little change with depth in a phantom exposed to X-rays.[4] Thus the glass reading can be taken as proportional to the actual exposure dose at the point in the X-rayed material with fair accuracy. Utilizing this conclusion we have made preliminary dose distribution measurements inside an X-irradiated mouse.

Two applications of the glass to dosimetry that seem most important to the author were investigated: (1) By extrapolating the reading per rad of glass plates of various thicknesses put in a β-radioactive solution to that of zero thickness, the absolute dose in the solution was measured. (2) By calculation from results using glasses of different silver concentrations, the thermal neutron fluxes and γ-contamination doses were separately measured.

II. MATERIALS AND METHODS

Silver-activated phosphate glass dosimeters were purchased from Bausch and Lomb Optical Co. Their elemental composition[11] by weight is $Al(PO_3)_3$, 46.3 per cent; $Ba(PO_3)_2$, 23.15 per cent; KPO_3, 23.15 per cent; and $AgPO_3$, 7.4 per cent. The glass rods are 1 mm in diameter and 6 mm in length while the glass plates are 1 cm square with thicknesses from 0.05 mm to 3 mm.

The measurement procedure followed was that of DEGELMAN *et al.*[6] except in the following particulars: (1) use of a single end-window photomultiplier (DuMont 6291) which gave sufficient stability for our purposes; (2) addition of an ultraviolet cut-off filter between the orange filter and photomultiplier to reduce background reading; (3) a rectangular slot in the rod carrier for the square glass plates; (4) 500-kΩ resistor and 0.1-μF capacitor as parallel input to 12AX7; (5) 5-kΩ instead of 500-Ω cathode resistors for 12AX7; and (6) smoothing capacitors across last stages of the photomultiplier. Fluorescence of irradiated glass rods or plates was measured between 3 and 24 hr after irradiation unless otherwise stated.

For all Co^{60} γ-ray irradiations in our experiments, the glass dosimeters were encased in plastic containers of 3-mm wall thickness. This encasing was done to approximate secondary electron equilibrium although theoretically a glass layer might have been preferable. Before measurement of the induced fluorescence, the glass rods or plates were rinsed in reagent acetone, washed in distilled water and finally dried with absolute alcohol. A clean glass surface is extremely important for good reproducibility. As the standard condition for reading fluorescence, the fluorometer was adjusted to give 102 μA for our standard rod which had been exposed to Co^{60} γ-rays about 6 months previously. Under this standard condition, the predose reading of glass rods from batch 1 was about 10 μA. The latter reading corresponds to the fluorescent output induced by about 77 r of Co^{60} γ-rays. The reading of a glass dosimeter for any radiation is obtained by subtracting the predose reading from the postdose reading.

III. RESULTS AND DISCUSSION

A. *Changes in fluorescent output of irradiated glass with time and temperature*

We checked the findings of SCHULMAN *et al.*[1,3] who reported that two opposing inherent factors affect the induced fluorescent output of the irradiated glass: one causing an increase and the other a decrease in the fluorescent output, and both being dependent on storage time and perhaps irradiation temperature. For example, we have observed a larger fluorescence yield at the irradiation temperature of 100°C than at room temperature, as found by SCHULMAN *et al.*[3] but the yields at 200°C and 250°C became smaller and smaller.

We found that the fluorescence reading increases with time after exposure and reaches a maximum value rather quickly, after which it decreases slowly. An example of such a change at room temperature is shown in Fig. 1. To avoid the error due to this growth of fluorescent output, we restricted ourselves to readings taken more than 3 hr after irradiation. We found the fading with time after this growth interval to be of the order of 5 per cent in the first month after irradiation when the detector is standardized by reference to a glass rod exposed 3 months before (Fig. 1). During the experiments, however, measurements of this reference rod showed a small increase in its fluorescent output compared to that of a manganese glass rod. If the latter output is assumed to be absolutely constant with time, the 5 per cent fading in a month is an overestimate.

Another important factor causing errors in glass readings is the temperature coefficient of

Table 1. Dose-rate dependence of fluorescence reading of silver phosphate glass for Co^{60} γ-rays

Glass	Irradiation facility	Dose rate (r/hr)		Reading/r* (arbitrary units)		Ratio of reading/r at high dose rate to that at low dose rate (%)
		High	Low	High dose rate	Low dose rate	
Rods, batch 1	USNRL	3.12×10^5	1.00×10^2	124.5 ± 1.2†	137.6 ± 2.1†	90.5 ± 1.8†
Rods, batch 1	Biology, ORNL	4.7×10^4	1.43×10^2	126.1 ± 2.4	135.2 ± 1.4	93.3 ± 2.1
Rods, batch 2	Biology, ORNL	4.7×10^4	1.43×10^2	136.0 ± 3.0	149.2 ± 8.4	91.2 ± 6.0
Plates $1 \times 1 \times 0.02$ cm	Biology, ORNL	4.7×10^4	1.43×10^2	2080.0 ± 42	2220.0 ± 44	93.6 ± 2.8

* All fluorescence readings of glass rods were taken under standard conditions (see Materials and Methods, in text) and normalized to those at 24 hr after irradiation using an empirical curve for the attenuation of fluorescence induced in glass with time after irradiation.

† Standard deviations for readings of from two to ten glass dosimeters.

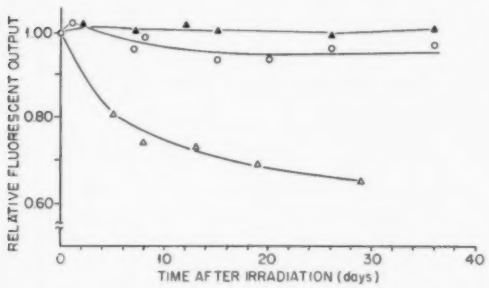

FIG. 1. Change of fluorescent output of γ-irradiated silver phosphate glass with time for various storage or irradiation temperatures. All rods were exposed to 330 r of Co^{60} γ-rays in 3 hr. The zero day reading was taken 3 hr after irradiation. ▲ = Glass rods irradiated at room temperature, stored at −5°C. ○ = Glass rods irradiated and stored at room temperature. △ = Glass rods irradiated and stored at 100°C (fluorescence read at room temperature).

fluorescence efficiency of irradiated glass, which is reported to be approximately 0·7 per cent/°C at 25°C.[1,3] A change of 1°C in room temperature has caused an easily detectable change in our fluorometer reading. To avoid this error, a glass rod exposed to Co^{60} γ-rays about 6 months before and fixed to the standard hole of the glass rod carrier was used as the standard rod throughout the rest of our experiments with rods.

B. *Dose rate dependence of glass reading for Co^{60} γ-rays*

Table 1 shows our experimental results obtained from the glass rods or plates exposed to Co^{60} γ-rays in the dose range 400 r to 6000 r. The significance of the results, of course, depends on the accuracy of the dose rate measurements of these γ-fields. For measurements of the two γ-field intensities in the Biology Division, ORNL, the same cavity ion chamber calibrated against a standard radium source has been used;[12] hence there will be only small errors in the ratio of the corresponding dose rates of the two γ-fields unless the ion chamber shows dose-rate dependence. This reasoning also applies for the γ-field intensities in U.S. Naval Research Laboratory and, in fact, the errors in the intensity measurements cannot be large enough to account for the observed dose-rate dependence.* The values obtained from glass rods of batch 1 exposed to different γ-fields, which had independently been calibrated by different methods, are in a good (±2 per cent) agreement for corresponding dose rates. This agreement in turn guarantees the accuracy of dose-rate measurements of the γ-fields and all figures for the dose-rate dependence are significantly beyond over-all experimental errors.

Therefore the induced fluorescent yield of

* ATTIX[13] exposed glass rods to γ-fields at the U.S. Naval Research Laboratory.

silver-activated phosphate glass depends on dose rate. The solid-state mechanism involved may be closely related to that causing the build-up or decay of fluorescence with time (Fig. 1). Although the glass measures radiation dose reproducibly, its accuracy may be improved by a more precise experiment on dose-rate dependence.

C. *Absolute measurement of absorbed dose in β-radioactive solution*

To compare quantitatively the β-particle effects on any material immersed in a radioactive solution with effects on the same material exposed to γ-rays, one must know the absorbed dose in the β-radioactive solution. Since glass response per rad in glass does not seem to depend on energy of photons,[9] we assume that β-ray response per rad is equal to the γ-ray response. Then, once the glass is calibrated by a known γ-field, we can measure the absolute absorbed dose in a β-radioactive solution.

Square plates with thicknesses 0.017, 0.031, 0.058 and 0.43 g/cm^2 were put in a P^{32} aqueous solution of initial specific activity 0.287 mc/ml $\pm$5 per cent for several hours.* The fluorescence reading per unit absorbed dose for zero thickness was estimated empirically by extrapolating the ratio of the fluorescence reading per unit thickness to the dose in rads in the solution, applying the method of least squares to the values of the three thinner sets of plates. Fortuitously, we had, as shown in Fig. 2, a perfect agreement of the extrapolated reading at zero thickness with the theoretical value. The latter was calculated from the Co^{60} γ-ray response per unit glass-rad of the plate, the exposure time, the specific activity of P^{32} and the mean β-particle energy. Despite the uncertainties in the calculations and extrapolation, the results demonstrate that the extrapolation glass-plate method is an easy way to measure with some accuracy the absolute absorbed dose in a radioactive solution.

From the practical point of view, however, it might be better to calibrate the fluorescence reading of a glass plate of finite thickness with a β-radioactive solution producing a known dose rate. Here we must know the amount of energy released in the glass plate which is calculable if we obtain the depth dose curve for glass plate of infinite thickness in contact with the plane surface of the β-radioactive solution.

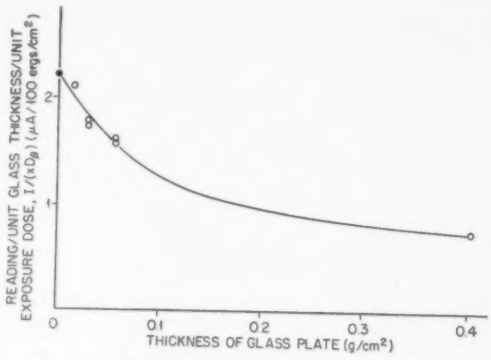

FIG. 2. Variation of fluorescence reading of silver phosphate glass plate placed in P^{32} aqueous solution with plate thickness. ○ = Experimental values. ● = Theoretical value calculated from the exposure dose in rads in the solution combined with Co^{60} γ-ray response of glass.

Imagine a β-radioactive solution in contact with an infinite plane surface of glass. Let $D(x)$ denote the dose in rad at the depth x(g/cm^2) in the glass system. We assume that LOEVINGER'S[14,15] empirical formula for $D(x)$ in tissue-like materials holds for the water-glass system. Then, the total energy, $U(x)$, absorbed per square centimeter of an infinitely extended glass plate, x g/cm^2 thick, immersed in the radioactive solution can be given by:

$$U(x) = 2\int_0^x D(x)\,dx = D_\beta\left[x - \nu x^2 \times\left(\frac{5}{12} + \frac{1}{6}\ln\frac{1}{\nu x}\right)\right];\ 0 < x < \frac{1}{\nu}$$

$$= \frac{D_\beta}{\nu}\left(\frac{11}{12} - \frac{1}{3}e^{1-\nu x}\right);\ \frac{1}{\nu} < x \qquad (1)$$

where D_β is the absorbed dose in rads in the interior of the radioactive solution and ν is a suitable absorption coefficient, taken as 9.1 cm^2/g.[14] Equation (1) ignores edge effects. In the limit of x tending to zero, $U(x)$ becomes

* Measured by ORNL Isotopes Sales.

equal to xD_β. This is the theoretical justification for our extrapolation method shown in Fig. 2.

The applicability of LOEVINGER's formula will be checked by the constancy of the ratio of the fluorescence reading $I(x)$ to the absorbed energy $U(x)$ calculated from equation (1). Table 2 shows the results of this numerical calculation together with the experimental data. In column 9 are shown the amount of deviation in per cent of the ratio $I(x)/U(x)$ from the fluorescence readings (per unit absorbed dose) of plates irradiated by Co^{60} γ-rays. The constancy of $I(x)/U(x)$ is not bad, but further improvement is necessary both experimentally and theoretically. For example, strictly speaking, the value of ν which we have used is valid only for tissue-like materials. Hence experimental determination of ν for our glass–water system is necessary.

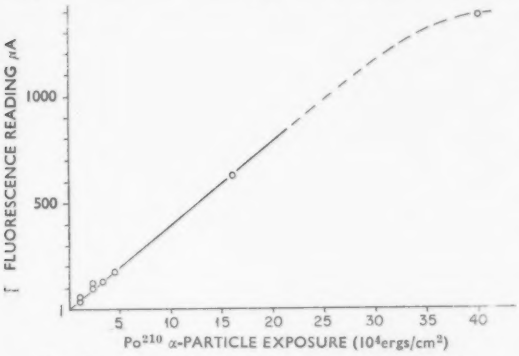

FIG. 3. Change in fluorescence reading of thin silver phosphate glass plate with α-particle dose.

D. *Response to α-particles*

Glass plates of 1 × 1 × 0.02 cm were exposed to α-particles from a thin layer of Po^{210}. There were roughly 1.7×10^6 α-particles cm^{-2} sec^{-1} on 11 September 1958.[16] The linear relationship between the glass reading and the absorbed dose is shown in Fig. 3. The fluorescence reading per rad in the glass plate for α-particles is only about 20 per cent of that for Co^{60} γ-rays.

E. *Responses of glass plates to* Co^{60} *γ-rays, 14-MeV neutrons and recoil protons*

Glass plates, 1 × 1 × 0.15 cm, encased in a lucite container of 3-mm wall thickness or covered with aluminum foil were exposed to 14.1-MeV neutrons as described previously.[17] As in the case of rods,[17] the higher response of lucite-covered plates compared with response of aluminum-covered plates is attributable to the contribution of recoil protons from the lucite container plus a minor contribution from heavy particles. By taking the difference of these responses we obtained the proton response of silver-phosphate glass. Comparison of these

Table 2. Relation of fluorescence reading to thickness of silver phosphate glass plates placed in P^{32} *aqueous solution*

Quantity	Glass thickness	Absorbed dose in solution	Calculated energy absorbed in glass	Observed glass reading	Normalized glass readings vs.			Relative discrepancy
					Solution dose	Energy absorbed	Dose absorbed from Co^{60} γ-rays	
Symbols	x	D_β	$U(x)$	$I(x)$	$\frac{I(x)}{xD_\beta}$	$i_\beta = \frac{I(x)}{U(x)}$	i_γ = calibration vs. Co^{60}	$\frac{(i_\beta - i_\gamma)\,100}{i_\gamma}$
Units	(mg/cm^2)	(10 rads)	(10^2 erg/cm^2)	(μA)	(μA/100 erg cm^{-2})			Per cent
Data	430	253 ± 13	250	775 ± 78	0.712	3.10	2.74 ± 0.08	+13
			325*			2.39*		−13*
	58	253 ± 13	106	235 ± 12	1.60	2.21	2.22 ± 0.09	−0.5
	31	511 ± 26	130.5	277 ± 14	1.75	2.12	2.22†	−4†5†
	17	511 ± 26	77.2	182 ± 18	2.10	2.36	2.22†	+6†

* These values include a 30 per cent correction for edge effects since the edge area was about 30 per cent of the side area. However, the correction to fluorescence reading should be less because fluorescence was measured only from the central area.

† Assuming that i_χ is the same as for 58 mg/cm^2-thick glass.

neutron and proton responses with Co^{60} γ-ray response is shown in Table 3. Calculation of the absorbed energy in glass for neutrons has been described[17] and that of energy released in the glass plates by recoil protons is given in the Appendix of this paper.

F. *Separate measurement of γ-contamination dose from thermal neutron fluxes*

Since any thermal neutron field is somewhat contaminated with γ-rays, knowledge of the γ-contamination dose is important for thermal-neutron irradiation of materials, especially biological materials. In this section we shall show how to measure separately the γ-contamination dose and thermal neutron flux in a thermal column. This method can, in principle, apply to any γ-contaminated thermal neutron field.

1. *Theory.* Let a phosphate glass dosimeter with x per cent $AgPO_3$ by weight, which is encased in a container of the same elemental composition so as to provide equilibrium of secondary electrons, be exposed to Φ thermal neutrons per cm^2 contaminated with Γ r of γ-rays.

Assuming the glass piece is thin enough that the first collision dose is applicable, the absorbed energy per gram, $D(x)$ rad, in the glass is given by

$$D(x) = D_n(x) + D_\gamma \tag{2}$$

where

$$D_n(x) = 10^{-2}\,\Phi \sum n_i \sigma_i E_{ij} f_{ij}$$

$$= \Phi[B(1 - x) + Ax] \tag{3}$$

$$D_\gamma = \frac{\Gamma}{1.18} \tag{4}$$

In equation (3), n_i is the number of the ith kind of nuclei per gram of the glass; σ_i the activation cross-section of nucleus i; E_{ij} the total energy released in the glass through j radiation emitted from neutron interaction with nucleus i; f_{ij} the average fraction of E_{ij} deposited in the glass; and ΦAx is the contribution from the $AgPO_3$ and $B(1 - x)\Phi$ is that from the remainder of the glass. In the derivation of equation (4) we have assumed that the γ-rays interact only by the Compton effect. Gamma-ray energies in thermal columns are usually high enough to justify this assumption. Inspection of the elemental composition of this glass immediately leads to the conclusion that D_n is mostly due to β-particles emitted from activated silver nuclei

Table 3. Fluorescence reading of silver phosphate glass plates, 1 × 1 × 0.15 cm, exposed to Co^{60} γ-rays, 14-MeV neutrons and recoil protons

Radiation	Material used to cover glass plates	Reading per unit exposure dose	Conversion factor from exposure dose to energy absorbed per plate	Reading per unit energy absorbed per plate (μA/100 erg)	Relative fluorescence reading
Co^{60} γ-rays	Lucite	1.02 (1 ± 0.03) μA/r	37.2† erg/r	2.74	1.00
14-MeV neutrons	Lucite	0.098 (1 ± 0.05) μA/Lucite rad	—	—	—
	Aluminum	0.072 (1 ± 0.03) μA/Lucite rad	7.30 ~ 14.6 erg/Lucite rad	0.98 ~ 0.49 (0.49 ~ 0.25)	<(0.18) >(0.09)
Recoil protons from Lucite	—	0.026* μA/Lucite rad	~4 erg/Lucite rad	0.65	0 24

Note: The values in the parentheses in columns 5 and 6 are corrected by taking account of γ-contamination.[17]

* This value was obtained by subtracting the value for aluminum-covered glass from that of Lucite-covered glass.

† This value was obtained from equation (4) in text using a glass weight of 0.43 g per plate.

and that the rest of D_n is due to β-particles and γ-rays from other elements.

As shown experimentally,[17] and as would also be expected since the fluorescent yield per glass-rad is independent of the energy of applied β-particles or γ-rays (see foregoing sections), the fluorescent output $I(x)$ of the irradiated glass is proportional to the absorbed dose $D(x)$ whether or not the thermal neutron field is contaminated with γ-rays; or

$$I(x) = C(x)WD(x) \tag{5}$$

where $C(x)$ is a proportionality constant which depends on concentration x of $AgPO_3$[1,7] and W is the weight. If the glass piece is given a known calibration dose D_0 rad of Co^{60} γ-rays, then

$$I_0(x) = C(x)\ WD_0 \tag{6}$$

where the dose rate of the calibrating and contaminating γ-ray fields should be comparable, or the dose-rate dependence discussed in Section B be taken into account. From equations (2)–(6) we obtain

$$\frac{I(x)}{I_0(x)} = ax + b \tag{7}$$

where

$$a = \frac{\Phi(A - B)}{D_0}\ ;\ b = \frac{D_\gamma + \Phi B}{D_0} \tag{8}$$

For calculation of A and B, we assume as previously[17] that in equation (3) the γ-ray contribution to D_n from activated nuclei of the glass is negligible because of small f_{ij} compared with $f_{ij} = 1$ for β-rays. Then, using the mean energy for β-particle emission from the activated nuclei multiplied by an appropriate factor and taking into account the half-life of the nucleus, if it is longer than the time between the observation of the glass reading and irradiation (see Ref. 18 for details), we can from equations (4) and (8) easily obtain

$$\Phi = 2.32 \times 10^8\ \Gamma_0 a\ (n_{th}\ cm^{-2}) \tag{9}$$

$$\Gamma = \Gamma_0(b - 2.7 \times 10^{-3}a)\ (r) \tag{10}$$

where Γ_0 is the exposure dose in roentgens of the γ-rays used for calibration.

If we plot the ratio $I(x)/I_0(x)$, i.e. the fluorescent output for a thermal column exposure divided by that obtained by a γ-ray exposure for calibration, against the $AgPO_3$ concentration x, then from equation (9) the gradient a can be converted to the integrated neutron flux Φn_{th} cm^{-2} and from equation (10), the intercept b and gradient a, the γ-contamination dose, Γ r, can be calculated.

2. *Experimental.* Glass plates of $AgPO_3$ added to a base phosphate glass [$Al(PO_3)_3$, 50 per cent; $Ba(PO_3)_2$, 25 per cent; and KPO_3, 25 per cent] by 8:100, 10:100 and 12:100 ratios by weight were obtained.[11] The 8 per cent $AgPO_3$ plates were $1 \times 1 \times 0.15$ cm and the two higher-concentration $AgPO_3$ plates were $1.5 \times 0.6 \times 0.18$ cm. Piles of four glass plates of each composition were exposed for 24 hr to thermal neutrons rather highly contaminated with γ-rays at the bottom of the water tank above the thermal column of the Oak Ridge Graphite Reactor.[19] Of the four plates of each group only the two lying in the middle were used, as a guarantee of equilibrium for secondary electrons.[17] For comparison, the neutron flux was independently measured by gold foils placed on both sides of the piles of glass plates. The glass plates of each composition were exposed for calibration to Co^{60} γ-rays of 259 r at dose rate of 112 r/hr at the Biology Division of Oak Ridge National Laboratory.

The results are shown in Fig. 4. As predicted by the preceding theory, the ratio of the response to the thermal column radiations and that to γ-rays only, $I(x)/I_0(x)$, increases linearly with increasing $AgPO_3$ concentration, x. Using the values for the 90 per cent confidence interval ($a = 9.4 \pm 3.4$ and $b = 0.74 \pm 0.37$) obtained from Fig. 4 by the method of least squares and the experimental value $\Gamma_0 = 259$ r, we obtain from the glass the values given in the first line of Table 4 where are also shown, for comparison, the values obtained through the courtesy of Mr. T. V. Blosser who used gold foils and an ion chamber, together with those obtained by Bagley *et al.*[19] several years previously. We expected that calculation from the glass dosimeter readings would give lower figures both for γ and thermal neutron doses because of the edge effects in the glass pieces used. The rather large deviations, by 10 per cent and 30 per cent, respectively, between Blosser's and our values of γ-contamination dose and thermal neutron flux

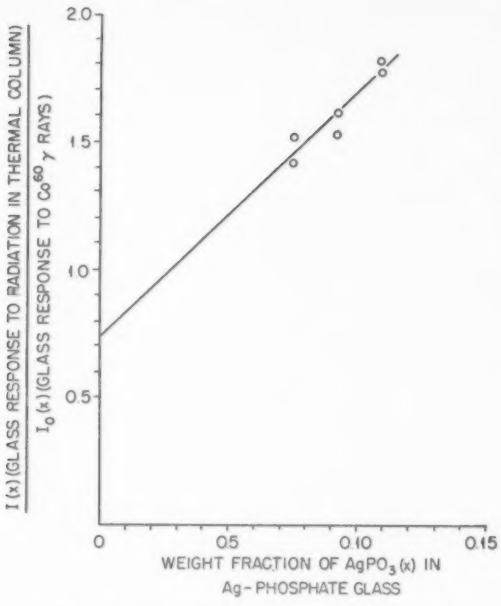

FIG. 4. Linear increase of fluorescence reading of silver phosphate glass irradiated in thermal column with increasing silver concentration.

found in these preliminary experiments may be because for each silver concentration we used only two pieces of glass, which showed large fluctuations in fluorescence readings. A later, similar experiment showed an improvement in precision and accuracy of the neutron flux and γ-ray dose values obtained.[20] Considering also the probable ± 10 per cent errors in the σ-values for silver nuclei,[21] the neglect of photoelectron contribution to the absorbed dose in the glass exposed to γ-rays, and the agreement of our values with the doses obtained by BAGLEY *et al.*[19] several years ago, we conclude that the results shown in Fig. 4 and Table 4 support the validity of the present method for coincident but separable measurements of γ-contamination dose and thermal neutron flux (see Ref. 18 for detailed discussion). Further improvement of this dosimetry method may lead to one of the simplest procedures to measure separately thermal neutron fluxes and γ-contamination doses with good accuracy.

G. *Dose distribution within an X-irradiated mouse*

The small size of the glass rods is an attractive feature of this dosimetry method for dose distribution studies.

For example, we made preliminary dose-distribution studies in a mouse irradiated with 250 kVp X-rays filtered by 3 mm Al, and found the dose to be constant to within ± 10 per cent in the spleen, liver, small intestine, kidney, testis and backbone. Mechanical or chemical action on the surface of the rods may affect the precision of such work.

The validity of using glass readings without corrections, in spite of the sensitive dependence of the glass reading on the X-ray energy,[1–3,7] has been checked by comparing readings of glass rods with those of a Victoreen 2500-r condenser chamber at various depths in a Lucite phantom exposed to the same X-rays. The results, plotted in Fig. 5, show good agreement between the two sets of readings. Such agreement has already been reported by ETZEL *et al.*[4] for other X-ray energy ranges. This agreement

Table 4. Comparison of thermal neutron flux (Φ/t) *and* γ*-ray dose rate* (Γ/t) *in thermal column estimated from fluorescence readings of silver phosphate glass dosimeters with* Φ/t *and* Γ/t *measured by gold foils and ion chambers*

Measuring method	Thermal neutron flux Φ/t (10^6 n_{th} cm^{-2} sec^{-1})	γ-ray dose rate Γ/t (r/hr)	γ-to-thermal-neutron ratio (10^{-10} r/n cm^{-2})	Observers
Glass	6.5 ± 2.3	7.7 ± 3.8	3.03	KONDO
Gold foil and ion chamber	9.3	8.5	2.54	BLOSSER
Gold foil and ion chamber	7.1	8.1	3.17	BAGLEY *et al.*

Note: Exposure time t for silver phosphate glass and for BLOSSER's gold foils was 24 hr.

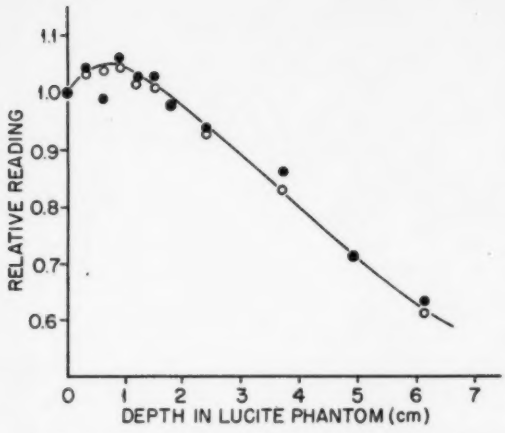

FIG. 5. Depth-dose curves in a Lucite phantom exposed to 250 kVp X-rays with 3-mm Al filter. ● = Fluorescence readings of silver phosphate glass rods. ○ = Readings of Victoreen 2500-r condenser chamber. Target-to-dosimeter distance, glass or Victoreen, was kept constant at 79 cm with varying distance between the surface of the phantom and the dosimeters. The Lucite backboard supporting the dosimeters was kept 3.7 cm thick.

may be explained by: (1) although fluorescence yield of the glass per roentgen depends sensitively on X-ray energy, the wide energy spectrum of X-rays generated by conventional machines may diminish the energy dependence of the glass, and (2) the energy spectrum of 170 kVp X-rays with 1-mm Al filter has been shown to change little with changing depth in Lucite of 5 cm thickness.[18] These observations encourage application of glass rods to internal dose measurement.

H. *Relative response of silver-phosphate glass to various radiations.*

The fluorescence readings of silver-phosphate glass per unit absorbed energy in glass are summarized in Table 5 for various radiations in units relative to that for Co^{60} γ-rays. Some of these data are preliminary. However, we may conclude that the fluorescence yield of the glass generally decreases with increase in LET value of the radiation. If the dependence of the fluorescence efficiency on the kind of radiation is only through the LET value, then one would expect the relative fluorescence reading to decrease gradually with increase of LET as is the case for the Fricke chemical dosimeter.[22] If so, our value for the ratio of proton response of glass to γ-response would be about 0.6; but the experimental value was 0.24. Thus we may conclude that fluorescence yield of our glass may depend not only on LET but also on the kind of applied radiation, provided that our estimated proton dose is correct. Hence, it is important to examine the proton response of glass for a wide energy range. There is a sharp contrast between variation of relative reading of glass with LET and that of RBE for chromosome aberrations.[10]

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IV. APPENDIX

An approximate estimate of the energy released in the glass plate, $1 \times 1 \times 0.15$ cm, by recoil protons from the Lucite container has been carried out for neutron irradiation as follows:

Table 5. Relative fluorescence readings of silver phosphate glass per unit absorbed energy in the glass for various radiations

Radiation	Relative reading	References
Co^{60} γ-rays	1.00	
Thermal neutrons	1.00 ± 0.10	Ref. 17
P^{32} β-rays	1.00 ± 0.10	Table 2
Protons recoiled by 14-MeV neutrons	$0.24 \pm ?$	Table 3
Po^{210} α-particles	0.20 ± 0.06	Fig. 3 and Table 2
14-MeV neutrons	0.14 ± 0.07	Table 3 and Ref. 17

Imagine a thick block of material with an infinite plane surface. Take the origin of x co-ordinates at the surface with the positive x-axis parallel to the inward normal and expose the block to neutron beams with energy E_n coming parallel to the x-axis from the positive side. Then, the energy dU released outside the material by recoil protons originating from the volume element by unit cross-section with thickness dx at depth x in the material is given by

$$dU = \nu_p dx \int d\Omega\, E(\theta,x)\sigma(\theta) \qquad (11)$$

where ν_p is the number of recoil protons per unit volume of the material; $\sigma(\theta)d\Omega$ the probability of protons recoiling into solid angle $d\Omega$ in the direction of angle θ with x-axis; and $E(\theta,x)$ the energy at the surface of the material of protons recoiled at x by neutrons in the θ-direction (laboratory co-ordinates).

Approximating the relation between maximum range R and the initial proton energy $E_i(\theta)$ by the conventional analytic formula, we have

$$R = a(E_i(\theta))^k = a(E_n \cos^2\theta)^k \qquad (12)$$

We now assume that a and k are constants, both independent of E_i. Then, from equation (12) we obtain

$$E(\theta,x) = a^{-1/k}\left(aE_n{}^k \cos^{2k}\theta - \frac{x}{\cos\theta}\right)^{1/k} \qquad (13)$$

From equations (11) and (13) combined with the isotropy assumption for angular distribution of proton recoils in the center-of-mass system, i.e. $\sigma(\theta) = \cos\theta/\pi$ in the laboratory system, we have

$$U = \nu_p \int_0^{\pi/2} \sin\theta d\theta \int_0^{2\pi} d\varphi \int_0^{aE_n{}^k(\cos\theta)^{2k+1}} dx \times a^{-1/k}\left(aE_n{}^k\cos^{2k}\theta - \frac{x}{\cos\theta}\right)^{1/k}\frac{\cos\theta}{\pi} \qquad (14)$$

This readily reduces to

$$U = 4R_0(1+k)^{-1}(5+2k)^{-1}k(\tfrac{1}{2}E_n\nu_p) \qquad (15)$$

Where R_0 is the maximum proton range for maximum energy E_n. Since the average energy of recoil protons is equal to $\frac{1}{2}E_n$, the factor $(\frac{1}{2}E_n\nu_p)$ in equation (15) is the energy imparted per unit volume of the material by $n-p$ scattering. Hence, equation (15) can be rewritten in the form

$$U = 4R_0(1+k)^{-1}(5+2k)^{-1}kf\rho D \qquad (16)$$

where D is the dose in rads in materials with density ρ and f the fraction of the total dose contributed by hydrogen.

For Lucite exposed to 14-MeV neutrons, taking $k = 1.8$, $R_0 = 0.18$ cm, Randolph's value $f = 0.634$[23] and $\rho = 1.18$ g/cm^3, we obtain

$$U = 4.0\,D \text{ (erg/cm}^2\text{)} \qquad (17)$$

where D is in Lucite-rads. Since our glass plates are thick enough to absorb all the energy of recoil protons from the Lucite container, the above value for U can be taken as the energy released in the glass plates by recoil protons.

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DOSIMETRIC PROPERTIES OF ELECTRETS

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Abstract—The dosimetric properties of resin–carnauba wax–beeswax electrets have been studied using γ-radiation from Co^{60}. The results are not completely accounted for by the explanation offered by MIAZDRIKOV in terms of the formation of pseudo dipoles within the dielectric. Attention is drawn to the practical disadvantages of electrets in dosimetry.

I. INTRODUCTION

ELECTRETS are generally prepared by allowing suitable molten dielectrics to solidify in an electric field. The resultant solid displays the property of a "semi-permanent" polarization which has excited rather desultory interest over many years. An account of early work on electrets is given by GUTMANN[1], while more recently the theory of the electret has been clarified by SWANN[2].

The electret is the electrical analog of the permanent magnet, and attempts have been made to apply its apparently attractive features in such devices as microphones, electrical meters, etc. In fact, BONING[3] lists many such devices in which the electret has been employed. Needless to say, however, widespread use has not been made of electrets, due in large part to certain practical disadvantages to which reference will be made below, with particular relevance to dosimetry. Nevertheless a patent has been issued recently in the U.S.S.R. for a dosimeter based on the use of the electret. An account of the theory and performance of this dosimeter is given by MIAZDRIKOV[4].

The Miazdrikov dosimeter consists of a flat circular electret of thickness Z_d lying on a conducting earthed plate. On the central part of the upper surface of the electret is laid a capped hollow cylindrical container of inside depth Z_g which thereby encloses a limited volume of air at some pressure P_g. For a surface charge density σ on the electret, an electric field exists through this volume of air. During irradiation, σ is changed due to two effects: the production of ions in the gas, which results in a current to the surface of the electret, and secondly, a leakage current within the electret also due to ionization.

Following the theories of GROSS and SWANN, MIAZDRIKOV develops the following expression for the rate of change of surface charge density:

$$-\frac{d\sigma}{dt} = cZ_g P_\gamma \frac{P_g}{P_0} \pm k\mu_{of}\mu_d Z_d I\dagger \qquad (1)$$

where c is the equivalent of the roentgen, P_γ is the magnitude of the radiation dose in the gas, μ_d is the linear absorption coefficient of the dielectric, μ_{of} is the moment of the pseudo dipole, P_0 is atmospheric pressure, P_g the pressure of the gas, and I the intensity of the incident radiation. The first term is the contribution of the ionic current in the gas, and the second is the contribution due to ionization within the dielectric.

The mechanism giving rise to the second term is rather complex. However, for the case of electromagnetic radiation of sufficiently short wavelength ($h\nu > 1$ MeV), the dominant effect is that of Compton scattering. Recoil electrons are scattered preferentially in a forward

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† Since surface charge density is equal to the normal component of the polarization per unit volume, the inclusion of Z_d would appear erroneous.

direction resulting in orientated, and relatively long-lived, electron-hole pairs, or pseudo dipoles. k is a factor which takes into account multiple production of pseudo dipoles by a single recoil electron. The choice of sign before the second term is determined by whether the orientation of the pseudo dipoles is such as to increase or decrease σ. According to MIAZDRIKOV, for electromagnetic radiation, the first term in (1) is always much larger than the second.

The present experiments were done without knowledge of MIAZDRIKOV's work, and the approach is different. The results, however, have a bearing on MIAZDRIKOV's theory. For electromagnetic radiation, the method employed by MIAZDRIKOV utilizes the field of the electret for the collection of ions formed in the air space above the electret and in this sense the dosimeter is similar to the condenser chamber. Our approach was to utilize the dielectric itself as the dosimetric substance, i.e. to make $Z_g = 0$ in the first term of (1). In this case only the second term of (1) should be operative and for electromagnetic radiation ($h\nu > 1$ MeV) the surface charge density should be linearly related to the time integral of the intensity of radiation.

II. METHOD

For the dielectric substance, a mixture, by weight, of 24 parts of resin, 24 parts of carnauba wax and 5 parts of beeswax was used. The mixture was brought to the molten state and poured into a cylindrical container with a brass bottom and Teflon wall. A brass plate was placed over the top, in contact with the dielectric, and a potential difference of 6000 V applied between top and bottom plates until the dielectric had solidified. Electrets of diameter 4 cm and thickness 0.5 cm were obtained.

Measurements of surface charge were made with electrostatic voltmeters of known capacity, and polarity of the surfaces determined with an electroscope.

Irradiations were performed with γ-rays from a Co^{60} therapy source. Except during measurements of surface charge density, each electret was kept tightly wrapped in aluminum foil at all times ensuring that $Zg = 0$ in (1). Measurements were made for several days prior to irradiation to make certain that the surface charge density of the electret had reached a constant value.

Irradiations were made for a period of 2–5 min, a measurement of surface charge density taken as quickly as possible, and the electret re-wrapped in foil and returned for irradiation. The uncertainty in the measurements is estimated to be less than 10 per cent. The effect of exposing the electret to the atmosphere during measurements was determined by monitoring other (non-irradiated) electrets simultaneously, and making the necessary corrections. This effect was found to be quite small— a reduction of voltmeter reading by only a few per cent for each exposure to the atmosphere.

In Fig. 1 is shown the reduction of surface charge density for a group of five electrets subjected to γ-radiation from a Co^{60} source. In Fig. 2, electrets A and B were irradiated together, with the negative side of A, and the positive side of B, facing the beam of quanta. Measurements in all cases were made with the positive side of the electrets resting on the grounded terminal of the voltmeter.

In Fig. 3 is shown the recovery of electrification of an electret after having been subjected to an exposure dose of approximately 2000 r from the Co^{60} source.

III. DISCUSSION

Whereas the theory of the electret dosimeter (sealed from the air) as given by MIAZDRIKOV

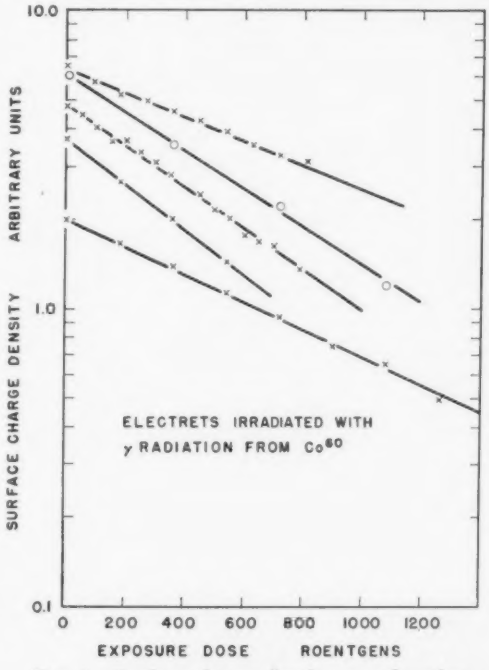

FIG. 1. Surface charge density as a function of exposure dose for a group of five electrets.

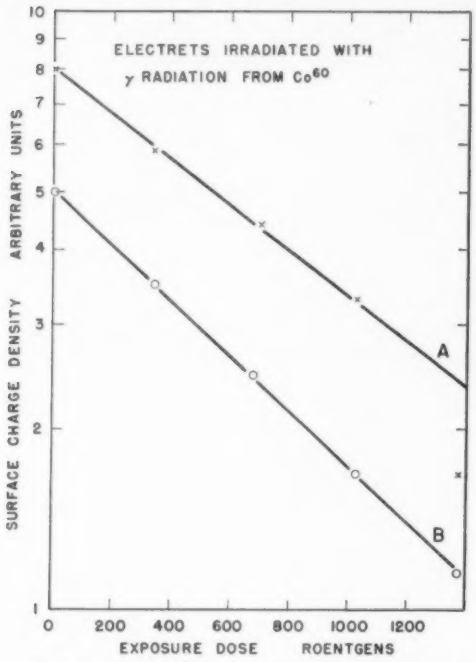

FIG. 2. Surface charge density as a function of exposure dose for two electrets, *A* and *B*, irradiated simultaneously. The negative side of *A*, and positive side of *B*, faced the beam of primary quanta.

requires that the surface charge density be linearly related to the exposure dose to which the electret has been subjected, the results shown in Fig. 1 would indicate that the surface charge density is more closely a logarithmic function of the exposure dose. Further the theory also predicts that the sign of the change in surface charge density will depend on whether the direction of flow of primary γ quanta is such as to give rise to a displacement of electrons toward the positive or negative sides of the electret. A comparison of curves *A* and *B* in Fig. 2 shows that there is a reduction in surface charge density regardless of which side (positive or negative) of the electret faces the direction of flow of quanta. It would appear therefore that the effects of irradiation on electrets are in general not completely accounted for by the theory of the formation of relatively long-lived pseudo dipoles within the dielectric. The contribution of these pseudo dipoles to the change in surface charge density seems to be masked by an additional effect. It may be argued that this additional effect is due to a layer of air, however thin, which exists between the electret and aluminium foil, i.e. that Z_g in (1) does in fact differ slightly from zero. It is to be noted, however, that in this circumstance the relationship between surface charge density and exposure dose would

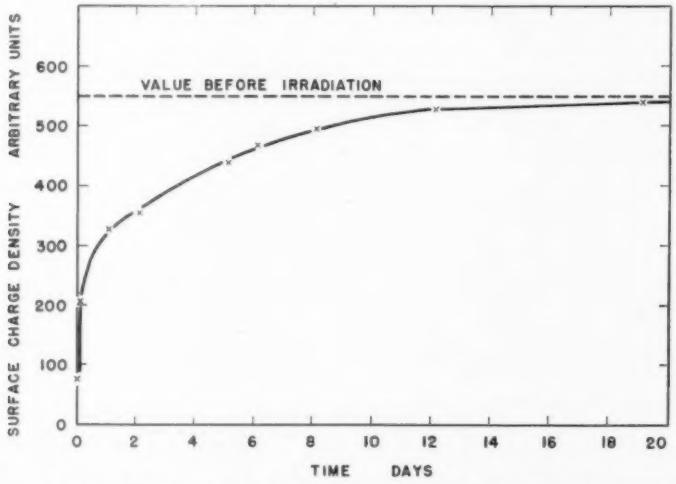

FIG. 3. Recovery of electrification by an electret after irradiation. The first measurement, at zero days, was taken immediately after completion of the irradiation. Except when measurements of surface charge density were being made, the electret was kept tightly wrapped in aluminum foil.

still be expected to be linear, as required by (1)*.

The results shown in Fig. 2 merely serve to emphasize the remarks made above with regard to the existence of an additional effect which masks the contribution of the pseudo dipoles to the change in surface charge density. There is no significant difference between curves *A* and *B*, and both are in fact similar to those shown in Fig. 1.

Fig. 3 shows that the reduction of surface charge density by irradiation is of temporary duration. In all irradiations performed, similar recoveries of electrification were observed.

This investigation was originally undertaken to determine the feasibility of using electrets as medium or high-level dosimeters. While the results would indicate that this is possible in principle, electrets have certain disadvantages to which attention must be drawn. The surfaces of electrets must be kept clean and dry; humid atmospheres must be avoided. The state of electrification is very sensitive to ambient temperature, at least for electrets of the resin–carnaube–besswax composition. Also it has proven very difficult to prepare electrets in a satisfactorily reproducible fashion, and finally, recovery of electrification after irradiation takes a considerable length of time. Of course, the possibility exists that electrets prepared of other dielectrics, and in other ways, may not be subject to these disadvantages, at least to the same extent. Failing this, however, widespread use of electrets (i.e. the dielectric itself) as a dosimetric substance would appear unlikely.

Acknowledgment—The authors wish to express their appreciation for helpful discussions with Mr. J. H. Aitken and Dr. W. R. Dixon.

* However, these experiments differ from those of Miazdrikov in that a conductor (aluminum foil) is in very close proximity to the surface of the electret. Accordingly the electric field between them is much more intense than exists in the Miazdrikov dosimeter, and the effect observed may in some fashion be associated with this greater field intensity.

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A COBALT NEUTRON-FLUX INTEGRATOR

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Abstract—The fast-neutron detector described here is expected to be most useful for health physics purposes. The detector is a cobalt disk embedded in a cadmium-cased paraffin moderator. The Co^{60} produced in a disk during irradiation is measured subsequently with a γ-ray scintillation spectrometer to determine the integrated neutron exposure. The 5.3-year half-life of Co^{60} permits accurate flux integrals to be obtained from exposure times as long as 1 year. The detector–spectrometer system can measure integrated exposures of 1×10^7 n/cm^2 or greater; the limit on the minimum detectable flux integral is imposed by spectrometer background count rate.

DESCRIPTION OF DETECTOR

The fast-neutron detector described here is simple and economical to construct, convenient and unobtrusive to expose at irradiation sites, and yields to relatively straightforward data analysis. It consists of a cobalt disk surrounded by a cadmium-covered hydrogenous moderator. The disk measures 2 in. in diameter by $\frac{1}{8}$ in. thick and weighs about 57 g. Paraffin is generally used as the moderator material, and can be in the shape of a 6-in. diameter sphere, a 6-in. cube, or a 6-in. diameter by 6-in. high cylinder. With such a moderator thickness, detector response is nearly uniform over a wide range of neutron energies, as shown by the work of Stephens and Smith[1]. The section view of a completely assembled detector appears in Fig. 1.

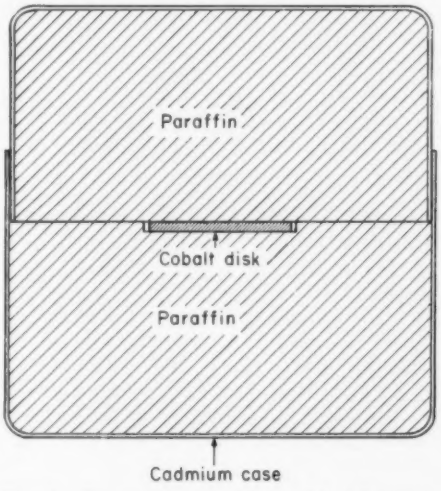

Fig. 1. Cut-away view of assembled detector.

Assembled detectors are located in a radiation field at stations where information on fast-neutron flux is required. Fast neutrons are moderated as they penetrate the paraffin, and those degraded to thermal energy at the cobalt disk are captured by the disk in the reaction Co^{59} $(n,\gamma)Co^{60}$. The Co^{60} content of the disk increases as the irradiation proceeds: in fact, the Co^{60} content is directly proportional to the time integral of the fast-neutron flux incident on the detector for exposure and decay times short compared with the 5.3-year half-life of this radioisotope. For example, the amount of Co^{60} present immediately after a 1-year irradiation in an arbitrarily varying flux will be within ± 7 per cent of the amount present from irradiation in a steady flux that delivered an equal integrated exposure.

Fig. 2 illustrates graphically the relationship among three flux integrals for different time

* This work was performed under the auspices of the U.S. Atomic Energy Commission.

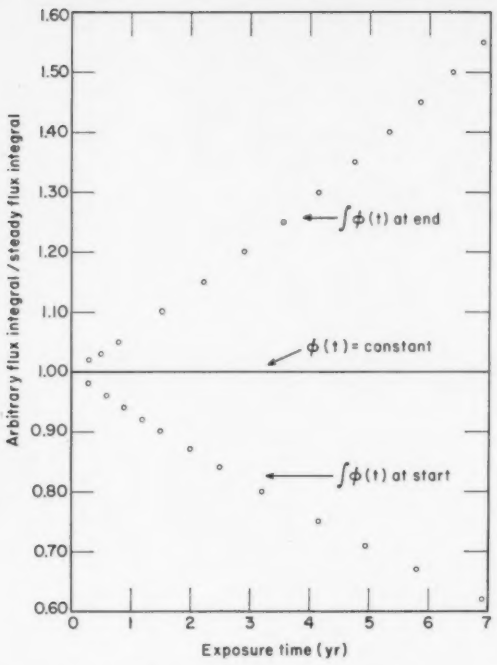

FIG. 2. Relationship among different time-dependent flux integrals.

intervals. These curves show the quantity of Co^{60} present at the end of an integration period, compared with the Co^{60} quantity present from an equal integrated steady-flux irradiation. The upper curve is for 100 per cent of the exposure occurring at the end of the irradiation time; the lower curve is for 100 per cent of the exposure occurring at the start of the irradiation time. The horizontal line at 1.0 represents the steady-flux irradiation. Any knowledge of the time variation in neutron-flux intensity will, of course, permit calculations of a more accurate value for the true integrated exposure. Therefore, the upper and lower curves on Fig. 2 represent the maximum possible errors that can result from unknown changes in irradiation rate.

DESCRIPTION OF ANALYZING EQUIPMENT

Analysis of the Co^{60} activity is performed with a γ-ray scintillation spectrometer.

Decay of Co^{60} is accompanied by the emission of two γ-rays in cascade, at 1.17-MeV and 1.33-MeV energy, respectively. These γ-rays are detected in a 3-in. by 3-in. NaI(Tl) crystal, coupled optically to a Dumont type-6364 5-in. diameter photomultiplier tube. Phototube output pulses are fed to a UCRL model-5 linear amplifier, which produces output pulses in the range from 0 to 150 V at an impedance level of $\sim$100 Ω. These pulses are coupled directly to the various analyzing units. The differential pulse-height spectrum is displayed on a fifty-channel pulse-height analyzer, also of UCRL design.[2]

Only those pulses that fall within the 1.17-MeV and 1.33-MeV photopeaks are accepted as Co^{60} decay events; in this manner the background (BKG) count rate is greatly reduced for a relatively small sacrifice in detection efficiency. One integral scaler is set at the lower edge of the 1.17-MeV photopeak; a second integral scaler is set at the upper edge of the 1.33-MeV photopeak. The difference between these two scaler totals equals the summation of all channels in the differential spectrum within the accepted energy interval. The two-scaler set provides a check on internal consistency of data and offers a possible alternate counting method if no multichannel differential pulse-height analyzer is available.

A third integral scaler is set at a threshold of $\sim$200 keV (lower edge of channel 5) to register a quantity that approximates the total scintillation events per run. A signal derived from the discriminator of this unit drives a binary-scaling count-rate meter, which in turn provides an input signal for the Leeds and Northrup chart recorder. Inspection of the chart record shows whether any significant change in count rate occurs during a measurement period.

The minimum integrated flux we can measure is limited by both the magnitude and constancy of BKG. Although considerable effort has been expended to reduce BKG to a constant low level, because of its present location the system is not usable during Bevatron operation and is noticeably affected by operation of the 184-in. cyclotron. Therefore we must know how BKG behaves during a low-activity determination. The scaler-chart recorder system detects primarily BKG when low-Co^{60}-activity runs are made. An average integral BKG count rate of $\sim$300 counts/min and the smoothing effect of cascaded binary-scaler stages combine to produce a relatively smooth chart-recorder trace. Thus, changes in BKG count rate of the order of 10 per cent can be seen easily if the changes persist for several minutes, i.e. long enough to perturb a measurement. The chart record has been of great value as a criterion for determining acceptance or rejection of low-activity counting data.

CALIBRATION OF DETECTOR

The detector has been calibrated with PoBe neutrons. A source distance of 50 cm and 166 hr irradiation time produced an integrated flux of 9.70 $\times$ 10^8 n/cm^2 at the detectors. Two cubical detectors were exposed—one with the plane of the cobalt disk perpendicular to rays drawn to the source, the other with the disk plane parallel to these rays—to test whether the detector showed any directional response characteristics, which are undesirable.

Results of the PoBe calibration are shown in Table 1. No significant difference exists between the count rates observed from the two disks in different orientations; therefore this detector is non-directional in its response characteristics. The third entry in Table 1 shows results obtained with disk 3 counted at a later date, and with a somewhat different BKG count rate.

Table 1. *Cobalt detector calibration,* PoBe *neutron data*

Disk number	Counts/min per disk from 1.0 $\times$ 10^7 n/cm^2	
	25 September 1959	2 October 1959
3	1.25 $\pm$ 0.03	1.22 $\pm$ 0.04
4	1.21 $\pm$ 0.05	—

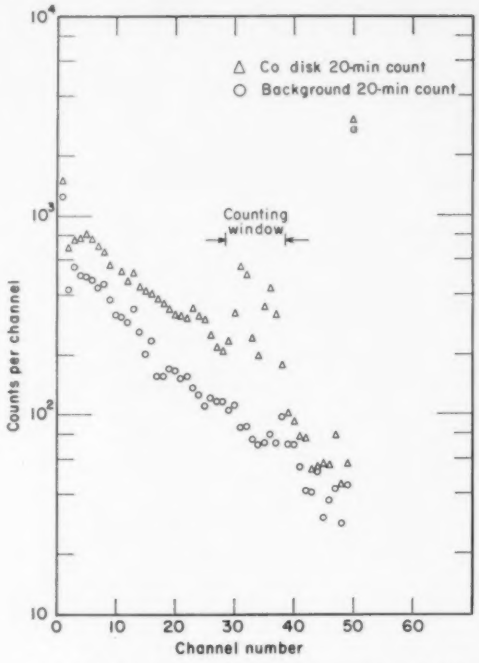

FIG. 3. Typical differential spectra for BKG and activated disk.

There is good agreement between these two disk 3 determinations. A typical Co^{60} spectrum and a BKG spectrum are plotted in Fig. 3. The Co^{60} spectrum is one of the calibration runs; the BKG spectrum is also from the calibration series. Both spectra are normalized to the same counting time. The counting-window limits are indicated by vertical bars on each spectra at the lower edge of channel 29 and the upper edge of channel 38. Note that in Fig. 3 the BKG count rate in the window is considerably above the normal value indicated below; 184-in. cyclotron operation produces this sort of effect.

The calibration constant of this detector for PoBe neutrons is 1.25 $\pm$ 0.03 counts/min per 1.0 $\times$ 10^7 n/cm^2 integrated flux incident on the detector. The listed error limits are those that derive from counting statistics alone, and are not meant to imply that our PoBe source strength is known so accurately. Source emission is probably known to within $\pm$ 5 per cent absolute. The BKG count rate in the window is nominally 17 counts/min. It is practical to measure 10^7 n/cm^2 with this system if the background can be kept constant throughout the counting time. For example, an integrated exposure of 10^7 n/cm^2 can be measured with a standard deviation of about 50 per cent in a 100-min count with the BKG at 17 counts/min. Efforts are currently directed toward decreasing BKG and increasing detection efficiency.

An unexposed disk, cut from the same stock as the calibration disks, was counted at this time. No Co^{60} activity above the 17 counts/min BKG was observed for a 10-min count. More information is needed about the Co^{60} activity in unexposed cobalt, particularly in view of the need to measure near-minimum flux integrals. We must not confuse any preexposure Co^{60} activity level with the activation produced during actual exposures. Work is in progress along this line.

Cobalt disk dimensions were chosen as a compromise among several factors. Both diameter and thickness should be large so that we can achieve a high count rate per disk from a given flux integral. Disk diameter should be small, however, so that assembled detectors are of practical size and weight, and so a reasonable-size NaI crystal can be used to count disks with good efficiency. The 2-in. diameter is an effective compromise here. Disk thickness was originally $\frac{1}{8}$ in. Experiments with disks $\frac{1}{4}$ in. thick by 2 in. diameter showed that this extra thickness

yielded only a 20 per cent greater count rate than $\frac{1}{8}$-in.-thick disks for equal flux integrals; self-absorption during activation had become the limiting factor for the thicker disk. Thus the $\frac{1}{8}$-in. value was accepted as the best disk thickness.

Increased count rate can be achieved if several disks are exposed simultaneously in separate detectors, and are counted stacked together on the crystal. A disk is found to give about 80 per cent of the standard position count rate when counted atop an unexposed disk. Progressively smaller but worthwhile increments are expected from the addition of several more disks in this fashion. For certain situations it may even be necessary to use larger-diameter disks (larger detectors) in combination with "parallel" exposure and "series" analysis in order to make successful measurements.

Increased counting efficiency can be attained if the cobalt disk is brought closer to the crystal surface. The crystal in use has an irreducible $\frac{3}{8}$-in. space between the rear crystal face and the closest disc position. A decrease of this space to about $\frac{1}{8}$ in. will be realized in a new Harshaw "matched window" unit which is on order; a detection efficiency increase approaching a factor of 2 may be achieved here.

A more sophisticated and very attractive low-background detection system is made possible by virtue of the Co^{60} γ-ray emission as a cascade process. Thus, a pair of crystals can be used with coincidence counting techniques. One proposed method is outlined briefly here. Scintillation events in the first crystal are measured by a single-channel differential pulse-height analyzer set to accept only the 1.17-MeV and 1.33-MeV photopeaks. Scintillation events from the second crystal are sent to the fifty-channel pulse-height analyzer, which accepts for analysis only those pulses in time coincidence with first-crystal photopeak pulses. An entire spectrum is displayed on the multichannel analyzer. The BKG can be virtually eliminated by the coincidence arrangement, and it is possible that the entire spectrum of the multichannel display can be accepted as valid Co^{60} decay counts. Some of the inevitable efficiency loss inherent in coincidence analysis can be regained in this manner. Detectors to implement this approach are also on order.

The energy response of the cobalt detector has not been experimentally investigated in detail. However, since the only essential difference between this detector and the indium foil detector described by STEPHENS and SMITH[1] is the substitution of cobalt for indium, the energy responses are expected to be very similar. Fig. 4 shows the measured energy response of an indium detector. This detector employs a 6-in.-diameter spherical paraffin moderator. The right-hand ordinate shows counts per minute per disk expected from cobalt for 10^7 n/cm^2 integrated flux of various-energy neutrons, normalized to the measured PoBe neutron point. Both energy response and calibration constant are expected to vary somewhat with detector shape. Thus the sphere will show enhanced response to less than 1-MeV neutrons compared with the cube, and the cube will show enhanced response to greater than 5-MeV neutrons compared with the sphere. The magnitude of change involved here, however, is small and can generally

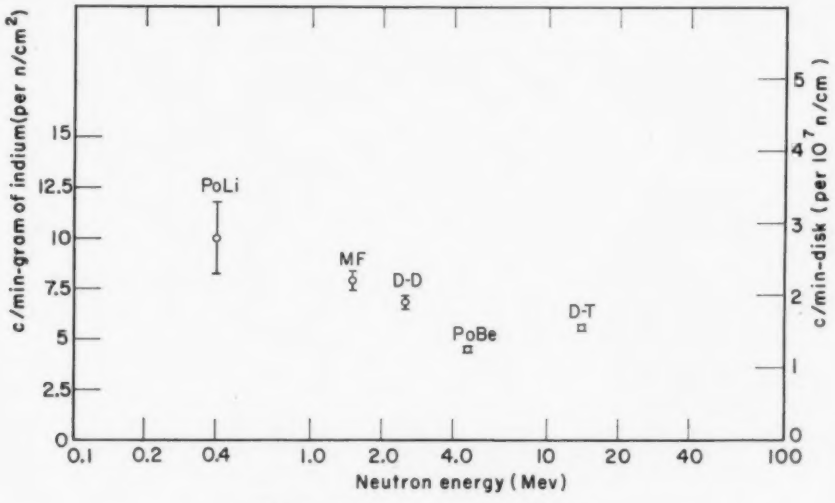

FIG. 4. Energy response of detector.

be neglected. The right circular cylinder was recently adopted as the standard detector shape at Berkeley. Additional calibrations at various neutron energies are scheduled, with emphasis placed on learning more completely the response characteristics of the cylindrical detector.

BACKGROUND-RADIATION PROBLEMS

Because of the great importance of the BKG count rate, short descriptions of the crystal detector, its enclosure, and associated BKG problems are included here. The NaI(Tl) crystal—a 3-in.-diameter by 3-in.-thick Harshaw unit—is cased in electrolytic copper with a fused quartz end window. The BKG observed with this crystal is significantly lower than the BKG observed with a second crystal of equal size but packaged in the conventional aluminum and glass container. Both crystal and phototube are housed in a light-tight iron box, which is buried inside a set of shields. A section view of the array appears in Fig. 5 with a cobalt disk shown in standard counting position. The 4-in.-thick Pb shield is primarily a γ-ray absorber. A 0.030-in. Cd sheet is provided to capture thermal neutrons; the outer 3-in.-thick paraffin shield moderates fast neutrons so that capture can occur in the Cd to prevent these thermal neutrons from reaching the crystal. (Boron should perform better than Cd in this capacity.)

We can prevent the acceptance of all scintillation events that occur during the actual acceleration cycles of such pulsed machines as the Bevatron and 184-in. cyclotron: the data recording units are electronically gated "off" for these periods. However, the residual BKG observed only during the "quiet" intervals is several times larger than normal BKG. The excess count rate was thought to be caused primarily by thermal-neutron activation of the crystal and nearby materials. To suppress this mechanism, Cd and paraffin were added to the Pb shield. A series of experiments was performed using the stray-neutron flux from the Bevatron (the most troublesome accelerator in the present context), which showed that crystal activation by thermal neutrons can account for essentially all the excess BKG, and that the Cd–paraffin combination is quite effective. Measurements with a Li^6I crystal show that the complete shield assembly provides an attenuation factor of $\times 32$ for the neutrons detected by this crystal. Unfortunately, the count rate above 200-keV energy threshold with complete shield and typical Bevatron operation is about 450 counts/min compared with the normal expected value of about 300 counts/min. The principal activity is definitely I^{128} with a 25-min half-life; therefore, relatively short-term changes in Bevatron operation can drastically change the BKG rate. In addition, sudden activation increases may accompany sample changing, which requires that the shield be opened. These effects, noted by observation of the integral BKG count rate, are reflected as equal or even greater relative changes of BKG within the Co^{60}-photopeak counting window. It has seemed too difficult to account accurately for such BKG fluctuations, and it has not been possible to move farther from the Bevatron (575 ft at present location). Thus the facility is used for low-level counting only when the Bevatron is off—approximately 10 hr per 168-hr week.

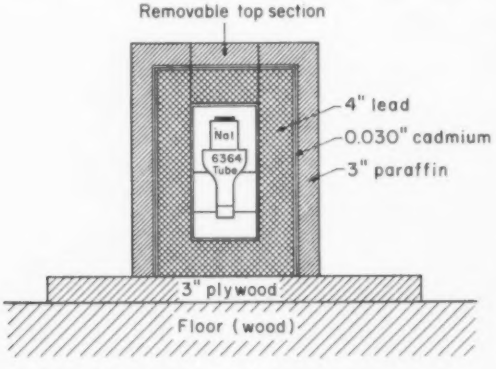

FIG. 5. Cut-away view of the crystal shield.

It is important to note that these difficulties are encountered in neutron fluxes at the shield exterior which are measured to be only: (a) 4.8 n/cm^2 sec slow-neutron flux (measured with a bare BF_3 proportional counter); (b) 2.1 n/cm^2 sec fast-neutron flux (measured with a BF_3 proportional counter encased in a paraffin moderator of such thickness as to exhibit nearly uniform response in the neutron energy interval from 0.1 MeV to 14.0 MeV). In particular, we

note that the thermal-neutron flux at the crystal is only about 0.15 n/cm² sec, and that this small flux induces a measured equilibrium I^{128} decay rate of about 150 counts/min in the 3 by 3 in. crystal.

As a matter of further interest, the cosmic-ray neutron flux at sea level reported by HESS *et al.*[3] can be described in the following manner:

(a) Total neutron flux intensity, including all energies: 0.04 n/cm² sec.

(b) Slow-neutron flux intensity, including energies from thermal to 30 eV, detected by bare BF_3 counter: 0.004 n/cm² sec.

(c) Fast-neutron flux intensity, including energies from 0.1 MeV to 14.0 MeV, detected by moderated BF_3 counter: 0.03 n/cm² sec.

If all item-3 neutrons were thermalized at our crystal, an elevation in the integral BKG rate of about 30 counts/min would be expected from I^{128} decay alone. In addition approximately 30 counts/min would arise from the activation process itself by detection of the capture γ-rays produced in the I^{127} $(n,\gamma)I^{128}$ reaction. Although this example represents the limiting case, it is possible that a significant fraction of the fast flux will be thermalized through an unfortunate combination of moderating and non-capturing materials, to become a potential crystal-activation threat. These remarks obviously apply to any other neutrons with similar energy characteristics. The results should be borne in mind when one attempts to perform careful low-level NaI(Tl) counting in the presence of *any* neutron flux appreciably greater than the natural BKG (cosmic-ray) flux.

APPLICATIONS FOR THE DETECTOR

For integrated fast-neutron irradiation the practical detectable minimum is defined here to be 10^7 n/cm²; there is no practical maximum detectable exposure. Accurate (± 7 per cent) flux integrals are obtained from 1-year exposure times. Therefore, the cobalt detector is potentially useful at any location where the flux integral is $\geq 10^7$ n/cm² year. Several combinations of exposure time and neutron flux to produce the 10^7 n/cm² minimum are plotted in Fig. 6.

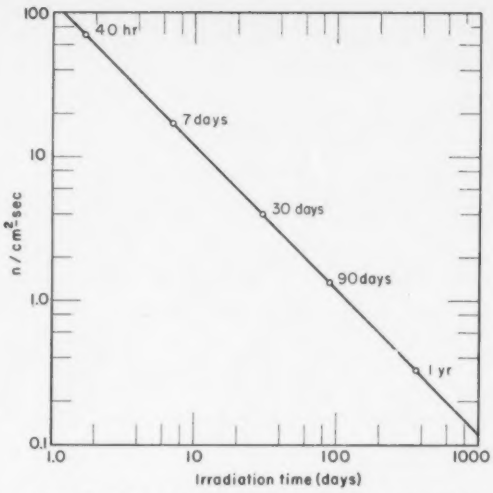

FIG. 6. Combinations of exposure time and neutron flux to produce the 10^7 n/cm² minimum detectable integral.

It is clear that the cobalt detector can provide neutron-exposure information for the low flux intensities of great interest to the health physicist. An important use for this detector can be to supply a record of the integrated flux at selected locations in the vicinity of neutron-producing particle accelerators and nuclear reactors. This record can be reexamined at leisure, and if properly stored in a low-neutron flux area, is a semi-permanent record (5.3-year half-life). In fact, any detector exposed to ≥ 10 rems of 1-MeV neutrons can be considered a permanent record, since the activity will be ≥ 1.5 counts/min after 25 years decay time.

Such a network of detectors is now in operation at L.R.L., Berkeley. The period of exposure is 30 days, chosen to coincide with the regular monthly exposures of both location films and personnel film badges. Detector stations have been selected on the basis that previous data showed these stations likely to be within the 10^7 n/cm² minimum per 30 days. Parts of many permanently occupied Laboratory buildings are suitable from this standpoint. Virtually all Laboratory buildings are suitable for the 90-day period; a schedule for certain stations in this category is being determined.

The integral flux value obtained from cobalt data can be converted to a biological-dose value (rem value) if the neutron spectrum is sufficiently well known. The experience gained at this Laboratory indicates that the neutron spectrum outside a well-shielded accelerator is relatively independent of both beam-target conditions and beam energy. Therefore, if special external beam situations are excluded, only the flux intensity and not the energy spectrum is expected to vary with accelerator parameters. A relatively few spectral measurements can then be used to provide factors for conversion of cobalt integral flux to rem values. A method used extensively at Berkeley for this purpose employs two counters exposed simultaneously to the neutron flux:

(a) Polyethylene-lined proportional counter with response proportional to the product of energy and flux density over the neutron energy range from 0.1 to 14 MeV.

(b) Moderated BF_3 proportional counter with response independent of neutron energy over the range 0.1 to 14 MeV.

The ratio of counts from these two counters is used to determine the mean neutron energy. This mean neutron-energy value is then used to determine the rem value appropriate to the flux integral obtained at that location.

Another application is in conjunction with, or in some cases as a substitution for, the "Oak Ridge type" threshold detector system recently adopted for potential criticality accident situations. The low cost of the cobalt detector, the ease of fabrication and exposure, the straightforward and unhurried analysis, and the permanence of record are advantages to be considered here.

The detector could also be used as a dosimeter for experiments in which integrated exposures of $>10^7$ n/cm^2 are delivered. The permanence of this detector record may be an advantage here; it may also be useful as a comparison against other dosimetry methods.

Problems such as standardization of counting techniques, calibration of detection systems, and intercomparison of results are expected to present only minor difficulties. In the first place, standard Co^{60} sources are readily available from the commercial market. Secondly, activated cobalt disks identical to those used in the detector can be produced easily by short irradiations in neutron beams of accelerators or reactors. And finally, particular activated disks can be sent around to all interested laboratories so that direct intercomparisons can be performed. The long half-life is advantageous for all these purposes.

PRECAUTIONS

Several notes of caution deserve mention in connection with the detector and its use. At least 1 hr should come between the end of a detector irradiation and the start of a cobalt-disk activity measurement. This period will allow for essentially complete decay of 10.5-min metastable Co^{60} (mainly into 5.3-year Co^{60}) and will also eliminate the presence of an unwanted 1.33-MeV γ-ray which accompanies the other branch (0.3 per cent probability) of 10.5-min Co^{60} decay.

If near-minimum flux integrals are expected, it is important to know the pre-exposure Co^{60} count rate of the disks. As a corollary, it may be important to store unexposed cobalt in a low-flux area; this should not be a difficult requirement. Storage of exposed disks may require a similar low-flux area if long-term data preservation is desired.

A potential difficulty with all activation detectors is the possibility that competing reactions will produce radioisotopes which decay in a manner that interferes with correct evaluation of the desired activity. The use here of a thermal-neutron reaction of relatively large cross-section (36 barns) tends to minimize the difficulty because all competing reactions are high-energy reactions with much smaller activation cross-sections. However, a low cross-section reaction that produces a short-lived activity can cause trouble. Therefore it is necessary to be particularly careful when analyzing disks that have been exposed in situations likely to produce such activities. The competing reactions generally have thresholds of from 8 to 10 MeV per nucleon or higher and are not expected to be troublesome outside well-shielded accelerators or reactors. However, exposure to the unmoderated fission spectrum

or to the energetic particles in unshielded locations at high-energy accelerators are potentially troublesome situations. For such cases it is important to examine the shape of the entire differential spectrum carefully, especially with regard to unwanted photopeaks at or above the Co^{60} peak positions. It may be possible to correct for the competing activities by a process of channel-by-channel subtraction of appropriate amounts of the pure γ-ray spectra from these isotopes. This technique is most successful when the unwanted isotopes are few, have simple spectra, and are available in reasonable purity as separate sources from which can be obtained the "master spectra" shapes required for the empirical subtraction process. However, the competing reaction products may be numerous, with resulting complex spectra. Then the most promising approaches would seem either to await sufficient decay of these products so that they can be ignored, or to make a detailed study of changes in the spectrum as decay proceeds. In general, disks that require the complex-analysis techniques will also show relatively high count rates, by virtue of the large exposures typically delivered in this sort of situation. Such disks will therefore make it possible to achieve the good statistical accuracy required for successful analysis by these methods.

DETECTOR FOR THERMAL NEUTRONS

The bare cobalt disk, without moderator or cadmium cover, can be used to integrate the thermal component of a neutron flux. The methods of analysis and precautions to be observed are identical to those which apply in the case of a fast-neutron detector. The activation produced by a thermal-flux integral is expected to be nearly the same as that produced from an equal fast-flux integral incident upon an assembled detector. The bare disk is therefore a very sensitive thermal-neutron detector from the standpoint of minimum detectable biological dose.

FUTURE PLANS

The direction of future effort has been indicated at appropriate places in the previous pages. In brief summary of the main items: efforts will be directed to increase counter efficiency, decrease counter BKG, and obtain good calibrations at several neutron energies. Particular importance is also placed on the coincidence-analysis technique. Progress along these lines, as well as representative data obtained from actual exposure periods, will be reported when such information is available.

CONCLUSIONS

The cobalt fast-neutron integrator is a simple and inexpensive detector which can perform accurate long-term flux integration. The detector sensitivity and the time-scale of activation or decay are such that accurate data can be obtained in the low fluxes of interest to the health physicist, and in practical time intervals. Furthermore, the data are faithfully retained, with a 5.3-year half-life, when proper storage precautions are taken. The detector could come into widespread use as the nuclear-energy field continues to expand and as greater importance is attached to the sort of information this device can provide.

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DOSIMETRY WITH SMALL SILVER-ACTIVATED GLASS RODS*

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Abstract—The use of small silver-activated phosphate glass rods for ionizing radiation dosimetry is described. Convenient methods for the handling, reading, and calibration of the rods are reported. With the apparatus described fluorescence of the rods may be determined with a precision of 4 per cent. With 20 MeV electrons and the γ-rays from radium, iridium-192, and cobalt-60, the smallest detectable absorbed dose is approximately 30 rads. With 250 and 180 kVp X-rays the smallest detectable dose is 3 rads. Variation in sensitivity of the rods with depth in an absorbing medium such as water is small with 22 MeV electrons, 250 kVp X-rays and 180 kVp X-rays incident on the surface. With the γ-rays from cobalt-60 a 40 per cent change in sensitivity was observed. The effects of exposure temperature, storage time, and ultraviolet exposure on the fluorescence of the rods are reported as well as the results of a limited number of applications.

INTRODUCTION

SEVERAL years ago SCHULMAN and ETZEL reported on the use of radiophotoluminescent glass in the form of small rods as a method of *in vivo* dosimetry.[1] The small size of these rods makes them very suitable for this purpose. Since the original report several papers dealing with the application of these rods to a variety of dosimetric problems have appeared.[2,3] Serious limitations to their routine use stem from difficulties associated with manipulation, reading, and calibration of the small rods, and their very large radiation quality dependence. In order to facilitate the routine use of these rods for *in vivo* dosimetry convenient methods of handling, reading and calibration of the rods have been developed. The results of calibration procedures, depth dose comparisons, and a limited number of applications are reported.

EXPERIMENTAL

Rods with optically polished ends, 6 mm long and approximately 1 mm in diameter, were purchased from the Bausch and Lomb Optical Company, Rochester, New York. The composition of the glass from which these rods were fabricated has been previously reported.[4] After sorting the rods into groups in which the diameter variation did not exceed ± 0.01 mm, they were stored in aluminum blocks (Fig. 1). From these blocks the rods could be conveniently transferred to the holder (Fig. 1). In order to measure the fluorescence this holder was inserted into a mounting block (Fig. 1) which could be positioned in a standard fluorometer (Photovolt Corporation, New York, model no. 540). When positioned in the holder the rod is framed by two 0.8 mm $\times$ 5.5 mm rectangular apertures at right angles, through which the exciting and fluorescent light pass. With this arrangement the light path through the rod is minimal and only a small portion of the rod volume is not used. The optically polished faces at the ends of the rod are shielded from both the exciting light and the photomultiplier. The fluorescence produced per unit energy absorbed (sensitivity) decreases at high fluorescence intensities (Fig. 2). The amount of this decrease is larger the longer the path length of the light in the rod. Since, in the experimental arrangement reported here, the exciting and fluorescent light directions are normal to the rod axis rather than parallel to the rod axis, the dependence of sensitivity on absolute fluorescence is much less than in the arrangement where the light enters the rod through its polished ends. A number 5860 Corning filter was used between the mounting block and the exciting lamp and a combination of Corning filters numbers 3484 and 4784 between the mounting block and photomultiplier tube. Rods may be inserted and removed with the aid of the pin in the holder as many as one-hundred times without affecting the fluorescence reading through chipping

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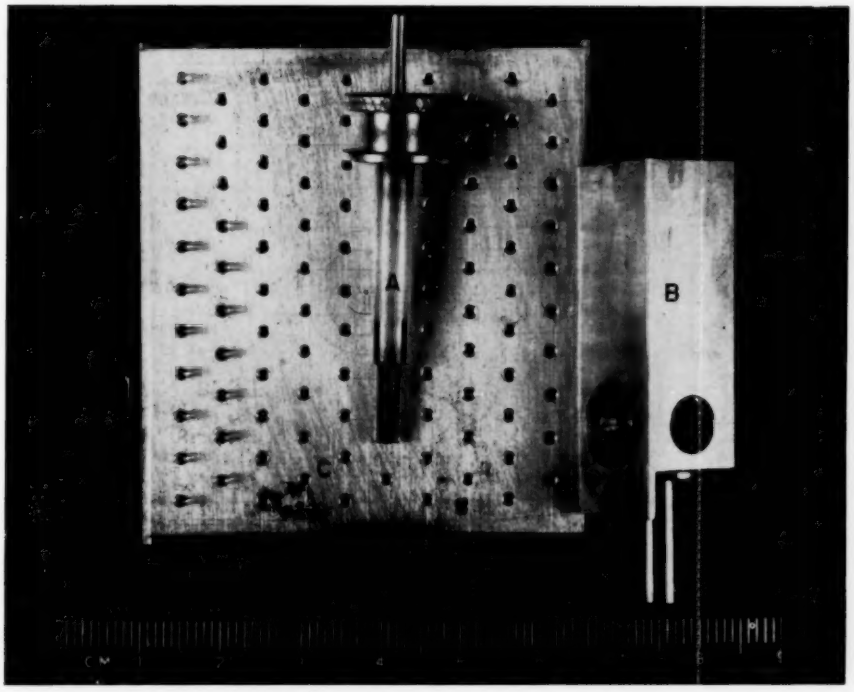

FIG. 1. Photograph showing holder (*A*), mounting block (*B*), and storage block (*C*).

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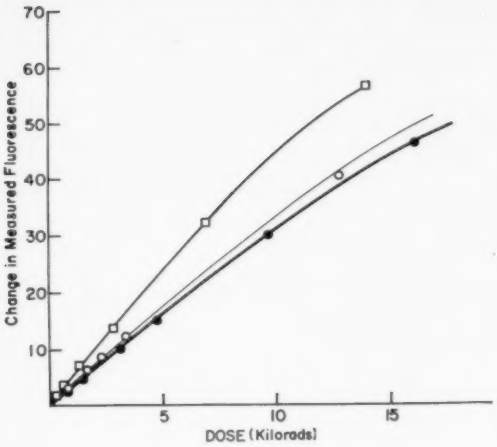

FIG. 2. Increase in fluorescence of rods as a function of energy absorbed in 0.8 N sulfuric acid surrounding rods. ●, Co^{60} γ-rays, 159 rads/min. ○, Ra γ-rays, 85 rads/min. □, Ir^{192} γ-rays, 680 rads/min.

and scratching. With this arrangement rods may be read by the operator at the rate of one every 10 sec. Positioning of the holder in the mounting block is very reproducible and leads to no detectable imprecision. The positioning of the rod in the holder is the principal source of irreproducibility as the rods are frequently "off-round".

Fluorescence of the rods was determined relative to the fluorescence of a manganese containing glass rod which was permanently mounted in a holder and used to adjust the sensitivity of the instrument. Once the sensitivity of the instrument was adjusted, it was stable over periods of many hours.

The rods were in all cases exposed to radiation in small cylindrical polystyrene containers filled with water. These containers measured 25 mm in diameter and 4 mm in height overall and had 1 mm thick walls. Energy absorption within the container was determined by exposing the container while filled with Fricke ferrous sulfate dosimeter solution. A *G*-value of 15.5 molecules per 100 eV was used to convert the measured change in ferric ion concentration to energy absorbed.

Calibration curves were obtained by exposing the containers filled with water and containing from one to three rods, 2 mm below the surface of a large tank of water and on the central axis of the radiation beam. In the depth dose studies the containers were arranged in a large tank of water at various depths below the surface and along the central axis of the incident radiation. The amount of energy absorption in the water immediately surrounding the rods was again determined by substituting Fricke dosimeter solution for the water in the container.

Rods used to measure skin doses were calibrated by exposing the rods while immersed in water $\frac{1}{2}$ mm deep contained in a cylindrical hole 2 cm in diameter on the surface of a polystyrene block. Energy absorption in the water surrounding the rods was again determined with the Fricke dosimeter.

RESULTS

Multiple readings of 500 individual rods indicated that the standard deviation of readings of an individual rod made on the same day was ± 4 per cent. This imprecision is associated with the positioning of the rod in the holder. The standard deviation of the readings of a series of different rods which had all received the same dose was ± 5 per cent, indicating that differences between rods in a selected diameter group (± 0.01 mm) are small compared to the positioning error.

Readings of individual rods from day to day had a standard deviation of ± 15 per cent, while the ratios of rod readings on different days had a standard deviation of ± 4 per cent. The data for rods exposed to 180 kVp X-rays are shown in Fig. 3. These data are typical of the results obtained for all radiations used.

Readings of unirradiated or "blank" rods indicated that the standard deviation of readings for a single rod was 5.4 per cent while the standard deviation of readings of different rods about a common mean was ± 5.8 per cent. From this it was concluded that the blank value for a group of rods could be satisfactorily obtained from readings of a single rod.

The effect of temperature on the sensitivity of the rods was investigated by exposing rods in water at temperatures from 24 to 64°C. The data for rods exposed to 250 kVp X-rays are presented in Fig. 4 and indicate an increase in sensitivity (fluorescence produced per unit energy absorbed in the Fricke dosimeter solution surrounding rod) of 0.4 per cent per °C.

Calibration results obtained with the various radiations used are indicated in Figs. 2 and 5. Data on the variation of sensitivity with depth are summarized in Table 1

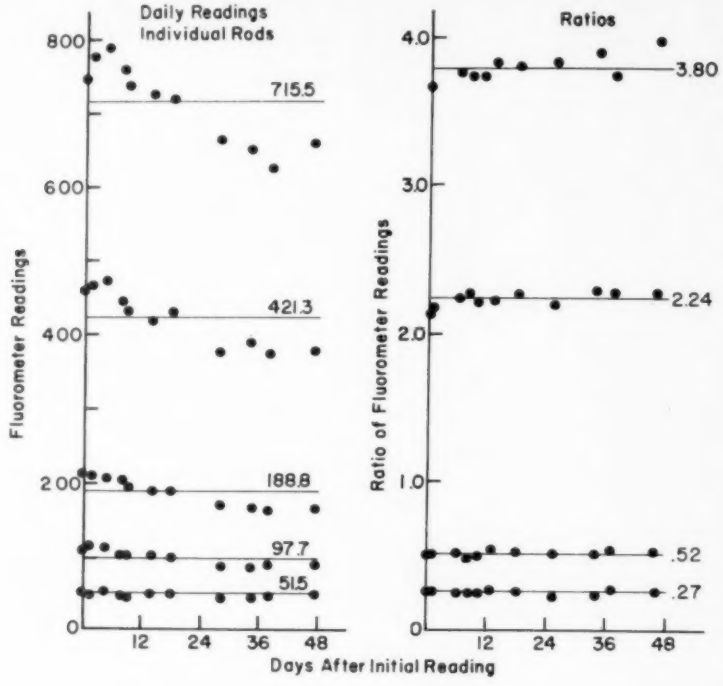

FIG. 3. Variation of absolute fluorescence reading and ratio of readings of rods irradiated with 180 kVp X-rays.

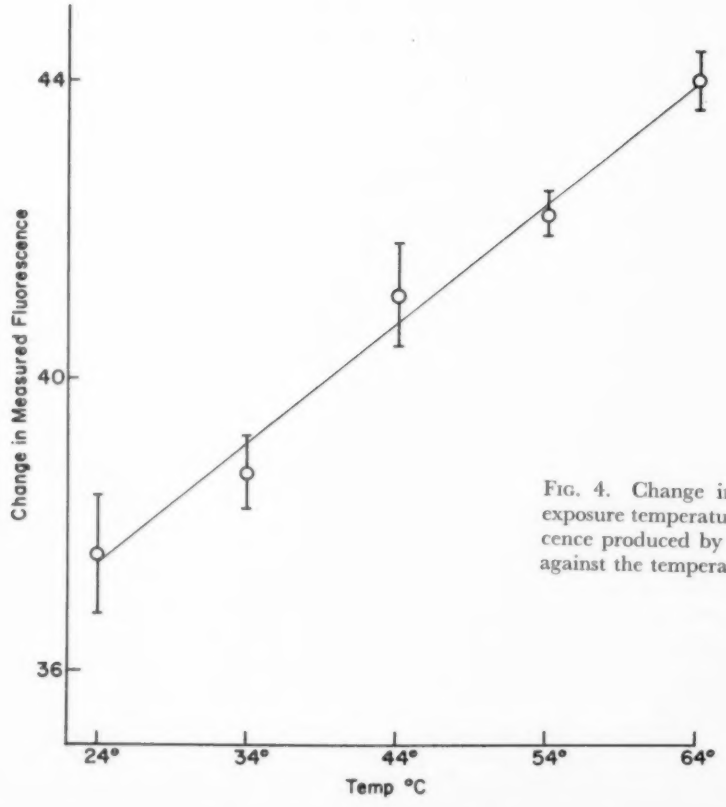

FIG. 4. Change in sensitivity of rods with exposure temperature. The change in fluorescence produced by a constant dose is plotted against the temperature during the exposure.

Table 1. *Sensitivity variation with depth along central axis in water for various radiations*

Radiation	Depth (cm)	Dose rate (rad/min)	(Sensitivity)$^{-1}$ [rad/(fluorometer unit)]
Co^{60} γ-rays	0.7	56.7	33.3
SSD, 50 cm	5.7	39.9	29.4
Field size, 25 cm	9.4	29.1	26.3
diameter at surface	13.7	21.3	25.6
	19.4	13.8	23.8
	24.2	9.7	23.3
	29.2	6.6	21.7
250 kVp X-rays	0	229	2.89
SSD, 40 cm	4.0	175	2.93
H.V.L., 2 mm^3	8.0	117	3.09
Field size, 30 cm	12	70.1	3.04
diameter at surface	16	44.2	3.19
	20	24.5	2.95
180 kVp X-rays	0	122	2.40
SSD, 40 cm	3	106	2.46
H.V.L., 1 mm^3	6	77.9	2.39
Field size, 30 cm	9	52.9	2.42
diameter at surface	13	33.4	2.49
	17	19.5	2.50
20 MeV electrons	0.4	102	42.9
SSD, 5 cm	2.6	99	44.3
Field size, 10 cm ×	4.8	84	42.6
10 cm at surface	6.7	65	44.1
	8.9	9.5	38.5

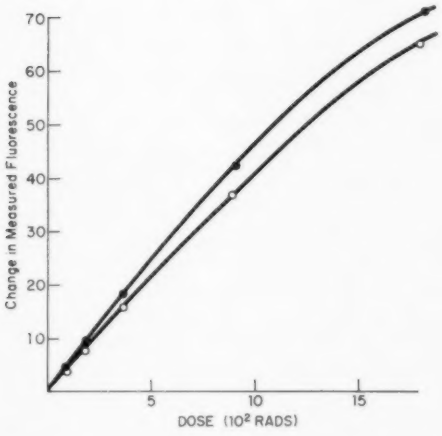

FIG. 5. Increase in fluorescence of rods as a function of energy absorbed in 0.8 N sulfuric acid surrounding rods. ●, 180 kVp X-rays, 180 rads/min. ○, 250 kVp X-rays, 177 rads/min.

The skin dose in a therapeutic procedure using 250 kVp X-rays was determined with the rods. These were affixed to the skin with scotch tape and had been calibrated as previously described. A Farmer–Baldwin intra-cavity chamber was simultaneously exposed and the chamber readings corrected to rads. The results obtained on two patients are shown in Fig. 6.

CONCLUSIONS

With the system of handling and reading the rods described here the sensitivity (energy absorption in 0.8 N sulfuric acid surrounding the rod per fluorometer unit) ranges from 3 rads/unit for 180 kVp and 250 kVp X-rays to 30 rads/unit for Co^{60} γ-rays and 20 MeV electrons. The calibration data summarized in Figs. 5 and 6 as well as the sensitivity variation in Table 1 are all directly comparable on the basis of energy absorbed in 0.8 N sulfuric acid surrounding the rod if it is assumed that the *G*-value of the Fricke dosimeter does not vary significantly for the radiations employed. We are at present in the

process of determining G (Fe^{3+}) for these radiations calorimetrically.[5] Ionization measurements made by others[6] indicate that G (Fe^{3+}) for 250 kVp and 180 kVp X-rays is a few per cent lower than the value used in this work.

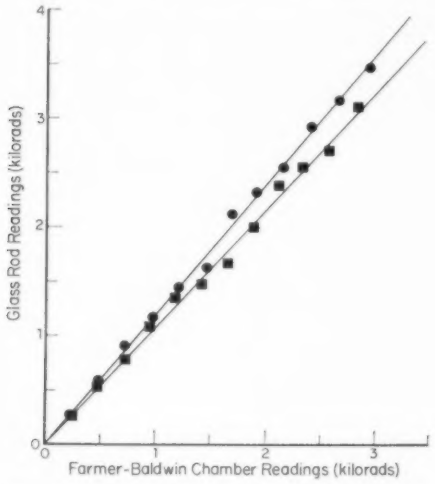

FIG. 6. Comparison of glass rod readings with Farmer–Baldwin chamber readings of skin dose. ●, S.L., slope = 1.19 ± 0.02. ■, G.S., slope = 1.08 ± 0.02.

At present the sensitivity of the rods is limited by the magnitude of the background fluorescence which is due to the fluorescence of Ag^+ centers. The magnitude of this background fluorescence could be reduced by decreasing the concentration of silver in the glass, and the resulting decrease in efficiency of fluorescent center production overcome by using greater detector sensitivity. A limited improvement in sensitivity might also be affected by sharpening the filtration of exciting and fluorescent light.

The 15 per cent standard deviation observed in absolute readings taken on different days is a reflection of the difficulty associated with reproducing the geometry of standard and sample and the small area of the rods exposed to the exciting light. Ratios of fluorescent intensity may be measured much more precisely (Fig. 3) and hence rods receiving an unknown dose when compared with calibrated rods may be used to determine doses absorbed in material surrounding the rod to within 5 per cent.

The relative fluorescent intensities of a series of exposed rods remains constant with storage time up to 6 months after exposure. The ratio of these intensities is independent of ultraviolet exposure for exposures corresponding to several hundred readings.

There is, however, a small fading of the absolute fluorescence with time which amounts to approximately 6 per cent per month. This was determined by measuring the ratio of the fluorescence of rods exposed identically but at different times. It is possible to apply a correction for this fading so that a given set of calibrated rods may be used for extended periods.

The sensitivity of the rods is dependent on the "quality" of the radiation and independent of dose rate. This pronounced "quality" dependence of the rods is the major deterrent to their routine use. It is possible to reduce this quality dependence by encasing the rods in high atomic number shields as has been suggested by SCHULMAN and also reported by MALSKY[7]. This increases their diameter significantly and lowers their applicability. With platinum shields designed and constructed at the United States Naval Research Laboratory and kindly loaned to us by Mr. FRANK ATTIX a significant reduction in quality dependence was noted. When calibrated with ferrous sulfate, as described above, rods encased in the shields were only 40 per cent more sensitive to 180 kVp radiation than to Co^{60} γ-radiation. The standard deviation amongst these rods rose, however, from 4 to 10 per cent. Studies on the origin of this imprecision are now in progress.

Rods containing significantly smaller amounts of high-Z material have also been tested by us. The quality dependence is reduced from a factor of 15 to a factor of 4 between 180 kVp X-rays and Co^{60} γ-rays.

The data of Table 1 indicate that the radiation sensitivity of the rods immersed in various depths of water along the central axis of 20 MeV incident electrons and 180 kVp or 250 kVp incident X-ray beams is constant within 4 or 5 per cent and that there is no detectable trend in the sensitivity down to depths where the dose rate is as low as 10 per cent of the maximum dose rate. Work is now in progress to determine the sensitivity variation off the central axis and as a

function of distance with the γ-rays from iridium 192 and radium isotope sources.

With Co^{60} γ-rays the central axis sensitivity variation is very different. There is a constant increase in sensitivity which amounts to approximately 50 per cent at a depth where the dose rate in water is 10 per cent of its maximum value. This change in sensitivity with depth severely limits the use of these rods with Co^{60} γ-radiation. Comparison of "depth dose" curves obtained with rods and ionization chambers[3,7] does not properly reflect this change in sensitivity with depth.

Acknowledgment—We are indebted to Mr. CHARLES STRYKER of our instrument shop for his invaluable aid in the design and construction of the mounting block and holder.

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PLUTONIUM INHALATION STUDIES—I

THE RETENTION AND TRANSLOCATION OF INHALED $Pu^{239}O_2$ PARTICLES IN MICE*

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Abstract—Mice exposed to $Pu^{239}O_2$ aerosols deposited about 20 per cent of the amount inhaled in lungs. From 40 to 60 per cent was distributed between the nasal and oral passages and the gastrointestinal tract. Although the major portion of the body burden was associated with the lung during a 70-week period there was gradual translocation to the skeleton, muscle, liver and spleen. The skeleton accumulated the largest quantity of translocated Pu^{239}, about 0.3 per cent of the total dose. This was equal to the amount remaining in lung. The half-life for clearance of about 4 per cent of the Pu^{239} initially deposited in lung was 460 days. The remainder was cleared with half-lives of 20 days or less. Assuming uniform distribution after deposition of 0.02 μc Pu^{239} in lung, the total α-dose to mouse lung tissue during the 70-week period was 550 rad.

INTRODUCTION

THE increasing availability of plutonium produced in nuclear reactors and its potential use not only in nuclear weapons, but as a reactor fuel, greatly increase the probability for human contamination. Experience has shown that a major route for internal contamination of humans by radioisotopes is by inhalation. Therefore, it is important to study the physiological behavior of inhaled plutonium and its biological effects for evaluation of the potential hazards of airborne plutonium particles.

In the present study mice were given a single exposure to air containing $Pu^{239}O_2$. Retention and translocation of plutonium were determined over a period of 70 weeks. The results provide some basis for evaluating the hazards associated with the inhalation of relatively insoluble plutonium dioxide.

METHODS

Exposure to $Pu^{239}O_2$ *particles*

Mice were individually placed head first into 50 ml conical centrifuge tubes with the bottom $\frac{1}{2}$ in. cut off to give a $\frac{3}{8}$ in. opening. The tubes containing the mice were inserted into ports drilled into the side of a vertical exposure chamber constructed of a 16-in. length of 8-in. (o.d.) lucite tubing. The exposure chamber held eighty-four tubes in seven rows of twelve each. The aerosol was admitted at the top and exhausted at the bottom through an electrostatic precipitator to collect plutonium and prevent contamination of the room air. The aerosol concentration varied approximately 25 per cent from the top to the bottom of the chamber. During all inhalation experiments aerosol samples were collected on millipore filters (type AA-Millipore Filter Corporation) and the particles characterized and sized by electron microscope examination. A more detailed description of the techniques used for exposing mice to $Pu^{239}O_2$ appears in a separate report.[1]

After exposure the 160 mice were divided, twenty to each stainless steel cage, and placed in temperature-controlled colony quarters maintained at 24°C. The mice were fed Purina Chow Checkers (Ralston Purina Company) and water containing 0.6 mg water-soluble A, B, D, E and C vitamins (Sharpe and Dohme Pharmaceutical Company) per ml. No deaths occurred during the 70-week period of observation and no significant gross pathology was observed when the mice were killed. Histopathology was studied in a parallel experiment and will be reported separately.

* Work performed under Contract No. AT(45-1)-1350 between the Atomic Energy Commission and General Electric Company.

Table 1. *Deposition and distribution after a* 2 *hr exposure to plutonium dioxide**

Experiment	μc Pu^{239}		Per cent of total inhaled†	
	A	B	A	B
Immediately present in GI tract	0.027 ± 0.014	0.067 ± 0.032	21	51
Immediately present in lungs	0.030 ± 0.0052	0.025 ± 0.0070	23	19
Immediately present in all other tissues	0.018 ± 0.013	0.0084 ± 0.0026	14	6.5
Total deposited	0.075 ± 0.014	0.10 ± 0.033	58	77

* Mean particle diameter by count was 0.2 μ.

† In both experiments the estimated total amount of plutonium inhaled by each mouse was 0.13 μc. This was based on assumed inhalation rate of 25 cm^3/min for each mouse.

Generation of $Pu^{239}O_2$ *aerosol*

A suspension of $Pu^{239}O_2$ (obtained from the Chemical Processing Department, General Electric Company, Richland, Washington) particles in 0.1 per cent Pluronics (polypropylene glycol–ethylene oxide polymer, Wyandotte Chemical Corporation) was placed in a small aerosol generator. Particles were dispersed in a fog by forcing air at 20 lb/in^2 through a 0.003 cm orifice at right angles to and just above the surface of the suspension. The fog containing $Pu^{239}O_2$ was then passed into the mouse exposure chamber. The particle size ranged from 0.05 to 0.6 μ in diameter. Both mean and median size by count were 0.2 μ. Ninety-nine per cent of the particles were 0.5 or less.* Generally the $Pu^{239}O_2$ particles appeared to be cubic in shape.

Tissue analysis

Groups of from ten to twenty mice were killed by cervical dislocation at intervals after exposure for determination of the tissue content of Pu^{239}. Tissues were removed and digested with nitric acid in small beakers. After ashing in a muffle furnace the residue was dissolved in 0.3 N HCl and the plutonium extracted with thenoyltrifluoroacetone. Aliquots were counted with an α-proportional counter. Tissues spiked with known quantities of Pu^{239} gave about 90 per cent recovery when digested and assayed in this manner.

RESULTS

Deposition and distribution following inhalation

The deposition and distribution of inhaled $Pu^{239}O_2$ particles in mice immediately following 2-hr exposures are shown in Table 1. The total microcuries of plutonium deposited (with standard deviations) and the per cent of the total plutonium inhaled are given. The total quantity inhaled was estimated from the concentration of radioactive particles in the exposure chamber, the duration of the exposure and by assuming an average minute volume of 25 ml of air per mouse.[2–4] The data are average values of from ten to fifteen mice killed immediately after exposure. In one experiment 21 per cent and in another 51 per cent of the total quantity inhaled was present in the gastrointestinal tract. About 20 per cent was present in lungs of mice in both experiments. Assay of all other tissues except skin accounted for 14 and 6.5 per cent in the two experiments. Almost all of this was associated with the head, and thus these values indicate the extent of the deposition in the nasal and oral passages. The total deposition in the two experiments was 58 and 77 per cent of the total inhaled. Thus, 42 and 23 per cent, respectively, were the fractions not deposited but exhaled. The seemingly poor agreement between these two experiments is not surprising since values for the total plutonium inhaled are estimates and not direct measurements.

* In more recent studies application of autoradiography to electron microscopy of particles collected on a millipore filter from a plutonium oxide aerosol showed the presence of non-radioactive particles. If non-radioactive particles were present in the plutonium oxide aerosols used in the above mouse experiments they were not distinguishable from the radioactive particles and were therefore included in the size measurements.

Table 2. Translocation of inhaled $Pu^{239}O_2$
(Per cent of total Pu^{239} *deposited—data are means of from fifteen to twenty mice)*

Time after exposure	Whole body	Lung	Bone* (total skeleton)	Muscle*	GI tract	Liver	Spleen	Kidney	Trachea	Ovary	Cervical lymph node and salivary glands
Immediately	100	33	8.8	4.9	52	0.22	0.070	0.052	0.64	0.0060	0.14
5 days	5.3	4.3	0.11	0.066	0.8	0.034	0.00023	0.0038	0.017	0.0021	0.0035
1 week	11	10	0.22	0.16	0.30	0.11	0.00071	0.011	0.031	0.00026	0.00069
3 weeks	3.6	3.2	0.070	0.027	0.046	0.040	0.0045	0.16	0.0047	0.00036	0.0015
5 weeks	5.1	4.8	0.19	0.060	0.040	0.030	0.0031	0.0063	0.018	0.00087	0.0010
9 weeks	1.4	1.3	0.060	0.013	0.019	0.0098	0.00050	0.011	0.0036	0.0012	0.00030
19 weeks	1.9	1.1	0.17	0.58	0.0086	0.020	0.0036	0.0026	0.020	0.0012	0.0023
51 weeks	1.3	0.84	0.22	0.019	0.0074	0.090	0.033	0.0060	0.011	0.00025	0.019
70 weeks	0.9	0.33	0.36	0.17	0.0060	0.016	0.011	0.00030	0.00010	0.00080	0.0030

* Includes upper nasal passages (skinned head) which received more than 14 per cent of the deposited dose. In other studies the skeleton and muscle excluding the head contained less than 0.1 per cent of the deposited dose. At later times the concentration of Pu^{239} in bone and muscle of the skinned head did not differ significantly from those of the rest of the body.

Translocation

To determine the translocation of Pu^{239} from lung to other tissues and the retention in those tissues, mice were killed periodically up to 70 weeks after exposure. The total original deposition was from 0.07 to 1 μc Pu^{239} per mouse. The distribution of Pu^{239} in several tissues is shown in Table 2 and is expressed as the percentage of the total Pu^{239} originally deposited. Immediately after exposure the gastrointestinal tract contained the largest percentage of deposited plutonium, 52 per cent, compared to 33 per cent in lung. The skinned head containing the nasal and oral passages was digested with the skeleton and muscle. This probably accounts for the large quantities reported for bone and muscle since there was no other evidence for rapid solubilization of such a large percentage of the deposited dose. As indicated in the footnote to Table 2, in another study when heads were digested separately, the combined bone and muscle contained less than 0.1 per cent of the deposited dose of plutonium. Immediately after exposure the liver contained 0.2 per cent of the total deposited and the kidneys, spleen, cervical lymph nodes and salivary glands contained less than 0.01 per cent. For the duration of the experiment, a major but continuously decreasing percentage of deposited plutonium was in lungs and associated lymph nodes. Bone contained less initially but showed increasingly greater amounts with time until the percentage in bone after 70 weeks equaled that in lung. There was also translocation of lesser amounts of Pu^{239} to muscle, liver and spleen. We cannot explain the inconsistently high value for muscle observed at 19 weeks. From about 10 to 70 weeks after exposure, translocation of Pu^{239} occurred within the mouse while the whole-body burden changed very little, from about 1.5 to 1 per cent.

For purposes of hazard evaluation the concentration in a particular tissue is more important than the total quantity because the absorbed dose of radiation is dependent upon the radioactivity per gram of tissue. This is particularly true of isotopes emitting α-particles with a range in tissue of about 50 μ. The concentrations of Pu^{239} for several tissues at various times after exposure are given in Fig. 1. A semi-logarithmic plot is used for graphic purposes only. Despite the translocation of Pu^{239} from lung to bone shown in Table 2, the concentration of Pu^{239} in lung was always greater by a factor of 100 at 10 weeks and a factor of 10 at 70 weeks. The concentration in liver, spleen and combined cervical lymph nodes and salivary glands paralleled that in bone. Ovarian tissue showed a higher concentration than bone. A similar observation was reported for $Ru^{106}O_2$ inhalation by mice, but the relatively high concentration in ovaries was not confirmed by autoradiograms

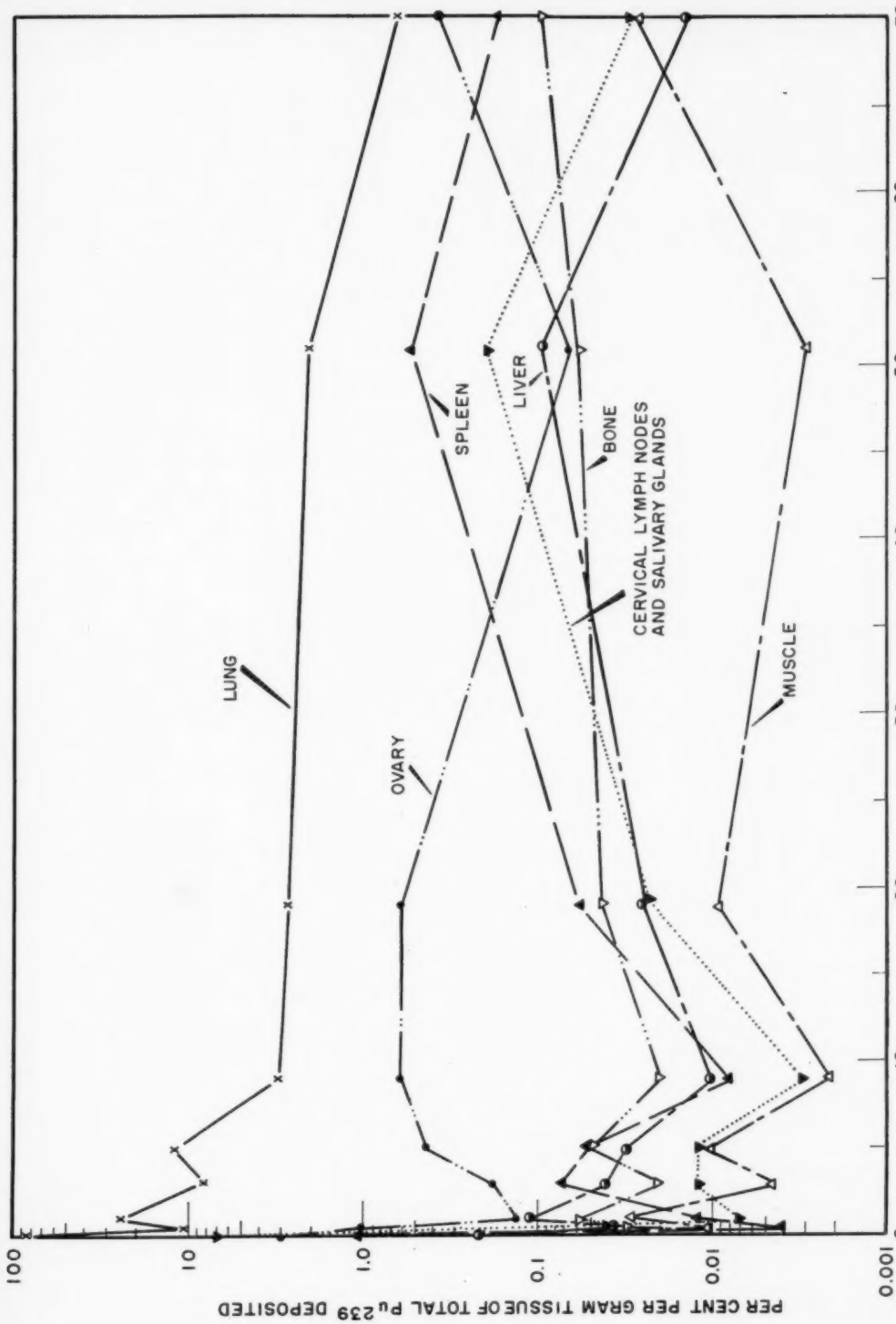

FIG. 1. Translocation of $Pu^{239}O_2$.

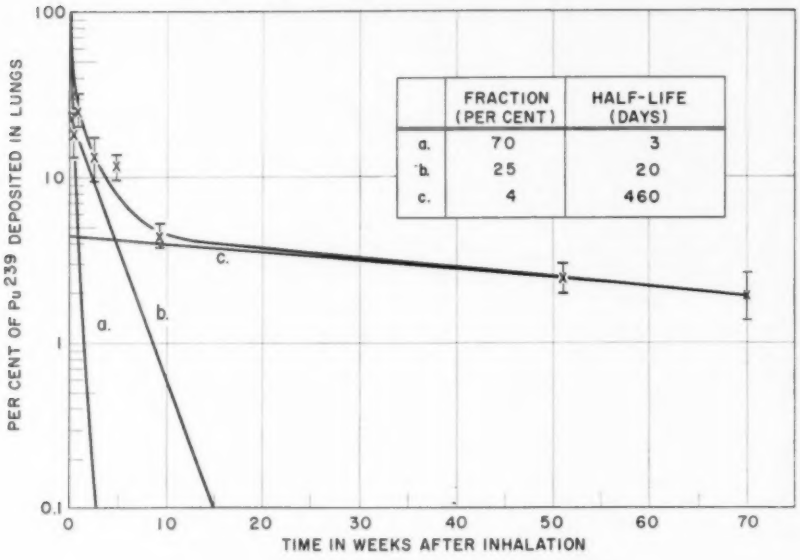

FIG. 4. Pulmonary retention of Pu^{239}.

(5). Similarly, in this study, the high concentration in ovaries was not demonstrable by autoradiography. However, only a few ovaries were prepared for autoradiograms.

Autoradiograms showed the presence of Pu^{239} in lung and lymph nodes. Figure 2 is an autoradiogram of a lung section removed 500 days after inhalation. The "α-stars" appear to originate on the walls of the alveoli. Figure 3 is an autoradiogram of a section from a bronchial lymph node from the same mouse showing α-tracks located throughout the section. The particles apparently are located in macrophages. A more detailed description of the histology associated with inhaled $Pu^{239}O_2$ will appear in a separate report.

Lung clearance

The clearance of Pu^{239} from lung and associated lymph nodes is of particular interest because these tissues maintained the highest concentration of plutonium throughout the experiment. Clearance was determined from the assay of lung and associated lymphatic tissue removed from mice killed at intervals after exposure. Lymph nodes, being very small, were digested with lung tissue. The data in Fig. 4 are expressed as per cent of the total Pu^{239} originally deposited in the lung. The standard deviation is given for each mean of fifteen or twenty mice. The clearance curve was resolved into three exponential components each having a different slope corresponding to the half-life of a particular fraction of the deposited material. The percentage of the total Pu^{239} originally deposited in the lung, p, that was present at a given time, t, is given by: $p = 70$ per cent $e^{-0.23t} + 25$ per cent $e^{-0.034t} + 4$ per cent $e^{-0.0015t}$. Biological (and effective) half-lives for the three fractions were 3, 20 and 460 days.

DISCUSSION

Deposition

The distribution of $Pu^{239}O_2$ particles in mice immediately after inhalation was similar to the distribution of $Ru^{106}O_2$ observed in another study.[5] As for Ru^{106} these data for Pu^{239} are also in the range of theoretical and experimental data summarized by HULTQVIST[6].

Translocation

Neither the translocation of $Pu^{239}O_2$ from lung nor the whole-body retention followed a

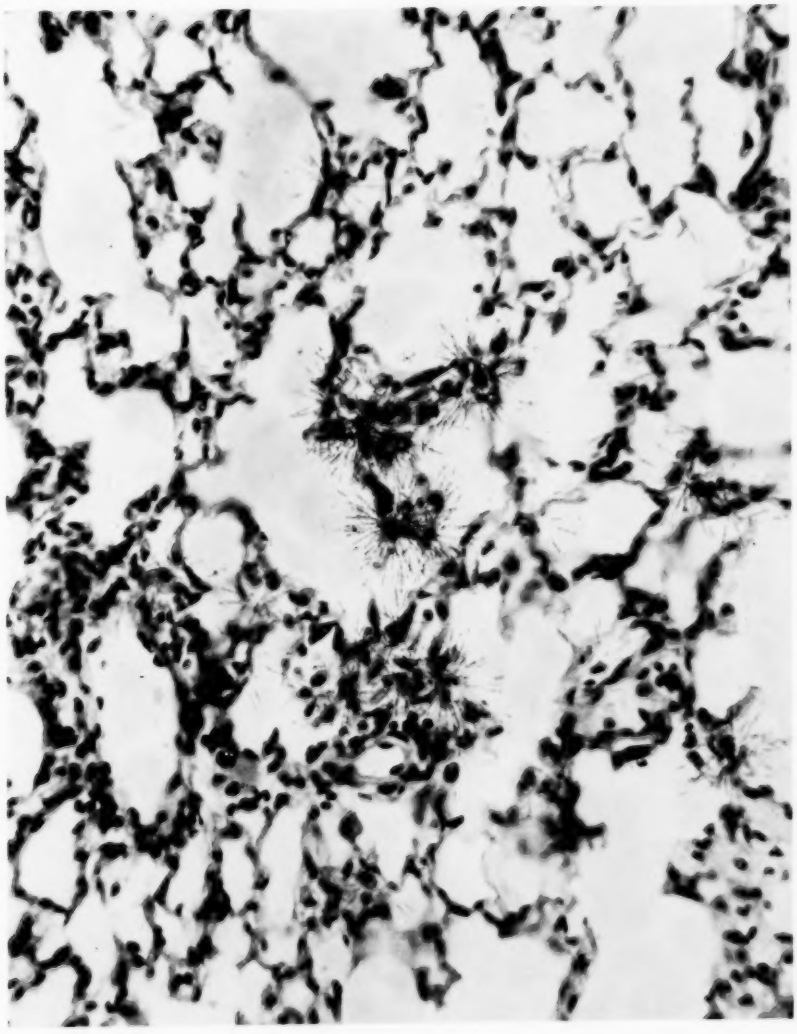

FIG. 2. Autoradiogram showing the presence of Pu^{239} in lung.

FIG. 3. Autoradiogram of bronchial lymph node showing α-tracks.

simple exponential relationship with time over the 70-week period. Because of the changing relationships between whole-body retention and lung burden there does not appear to be an easy way to predict lung burden from whole-body retention or total excretion of Pu^{239}. The translocation of plutonium to the skeleton was qualitatively similar to that reported by ABRAMS *et al.*[7] and discussed by LANGHAM[8] for mice after inhalation of $Pu^{239}O_2$ fumes.

The relative concentrations of Pu^{239} in tissues other than lung are not in disagreement with the distribution seen after intravenous injections of plutonium. KAWIN and DOCKUM[9] found high concentrations in bone and ovary with less in liver and kidney, 40 weeks after intravenous injection of citrated plutonium. Thus, in these studies there is no evidence that Pu^{239} translocated from lung and associated lymphatic tissue behaved differently than Pu^{239} given intravenously.

Lung clearance

The combined lung and associated lymph nodes received the greatest total radiation dose because they maintained the greatest concentration of plutonium throughout the experiment. An estimate of this dose is of interest in explaining possible pathology being studied in a parallel experiment. This estimate may be obtained from the clearance data with the necessary assumption that the Pu^{239} was uniformly distributed throughout lung and lymphatic tissue although the autoradiograms (Figs. 2 and 3) showed the Pu^{239} to be present in particulate sources. Using the exponentials for clearance the estimated total energy absorbed in lung and lymphatic tissue during the 70-week period following deposition of 0.03 μc $Pu^{239}O_2$ in a mouse lung was 550 rads.

Application to derivation of maximum permissible limits

Because of the interest in maximum permissible limits, the data obtained in this study were used to derive a value for the maximum permissible concentration of $Pu^{239}O_2$ in air. It is assumed that plutonium was uniformly distributed in lung tissue.* Ignoring other anatomical and physiological differences the extrapolation from mouse to man is on the basis of differences in organ weight.

The maximum permissible lung burden, the quantity at equilibrium resulting in a mean dose rate of 0.3 rem per week to lung, is estimated as 0.015 μc Pu^{239} using the equation and data given by the ICRP Committee II on Permissible Dose for Internal Radiation.[10] The concentration of $Pu^{239}O_2$ in air breathed 8 hr/day, 40 hr/week, over a period of 50 years resulting in an equilibrium lung burden of 0.015 μc is calculated using pulmonary clearance data from these experiments. The method of calculation was described in a previous paper (5). The value for the MPC_a obtained in this manner is 1.6×10^{-10} $\mu c/cm^3$ of air. Thus, assuming the behavior of plutonium in man is similar to that in mouse, the value given by the ICRP for insoluble plutonium of 4×10^{-11} $\mu c/cm^3$[10] may be on the conservative side by a factor of 4. Other experiments are in progress with other species to obtain more confidence in the extrapolation of results of radioactive inhalation studies from laboratory animals to man, and to determine the extent of accumulation of plutonium in lymph nodes.

Acknowledgments—We are grateful to M. G. BROWN for assisting in performing the experiments. We are also indebted to the following personnel of the Biologicial Analyses Operation: B. J. MCCLANAHAN and A. C. CASE for radiochemical assays; Dr. R. BORASKY for electron microscopy; and E. JANE COLEMAN and N. L. DOCKUM for photomicrographs and histology. Dr. H. A. KORNBERG's contribution of encouragement and advice are particularly appreciated.

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* For these calculations lung tissue necessarily includes associated lymph nodes because they were not analyzed separately for plutonium in this experiment.

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SOME CALCULATIONS ON THE ACCEPTABLE LIMITS FOR THE INHALATION OF RADIOSTRONTIUM IN AN EMERGENCY

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Abstract—Acceptable limits for the inhalation of radiostrontium in an emergency are calculated. Several methods are used to calculate the acceptable limits based on the body burden concept, the maximum allowable dose to the whole skeleton or the maximum allowable dose to a particular site in the bone. The authors conclude that 0.5 μc is an acceptable amount for inhalation by adults of strontium-90 and 16 μc for strontium-89 in conformity with the accepted levels for the permissible dose in bone. A short discussion is given on applicability of these values to children in the environmental population.

INTRODUCTION

THE exposure of environmental populations to an aerosol containing radiostrontium might occur as a result of a reactor accident. There is great difficulty in deciding the acceptable inhalation levels to which these populations may be exposed in an emergency. The report issued by the ICRP[1] after their Munich meeting states that the subject was extensively discussed and it was considered that the British Medical Research Council's Committee Report[2] was a useful and sound approach to the subject. A possible alternative method of calculating the acceptable inhalation level is to combine the two ICRP concepts of allowing a 13 week permissible intake to be accumulated in a short period of time (para. 52(e)). and permitting the environmental population to acquire one-tenth of the occupational exposure (para. 56).

The MRC Committee recommended that the emergency maximum permissible dietary intakes for strontium-89 and strontium-90 for the environmental population should correspond to a total dose of 15 rads and an annual rate of 1.5 rads per year, respectively, at the sites of highest concentration in bone, as compared with an occupational rate of 15 rads per year in bone. It should be noted that this is different from the ICRP recommendation of a mean dose to bone of 30 rems per year which is equivalent to 6 rads per year when the relative damage factor of 5 for β-emitters is applied.

In considering dietary contamination, the MRC Committee commented that if the daily diet contains 2 mμc of strontium-90 associated with a normal intake of 1 g of calcium, then the strontium-90 concentration at the sites of new bone formation cannot exceed one-quarter of this dietary ratio, or 0.5 mμc/g of calcium. This reduction of the dietary ratio of strontium to calcium results from the differences in the absorption and excretion of the two elements and has been reported by ECKELMANN[3] and BRYANT *et al.*[4] and other workers. The dose rate in bone which has been deposited with a uniform concentration of 0.5 mμc/g of calcium is a little below 1.5 rad per year. Similar considerations for strontium-89 show that a dietary intake of 200 mμc daily will give rise to a dose not exceeding 15 rads to the sites of highest concentration in the bone.

If the same approach as used in dietary contamination is used in the problem of inhaled radiostrontium then the following interpretation could be made. Assuming that all the inhaled strontium is in soluble form then 0.4 of the inhaled activity passes into the blood according to the ICRP standard man.[5] The blood of an adult contains about 300 mg of calcium[6] so that if the activity in the blood due to a short

period inhalation of strontium-90 is 0.15 mμc then bone deposited with this ratio, i.e. 0.5 mμc/g of calcium, will receive a dose not exceeding 1.5 rad per year. The results of animal experiments described by LANGHAM[7] show that the transfer of most of the radiostrontium from the lungs to the skelteon via the blood takes place in 2 or 3 hr. If the amount of strontium-90 reaching the blood is 0.15 mμc then the activity inhaled is 0.15 mμc/0.4, i.e. about 0.4 mμc. This is a very low value for permissible inhalation of activity in an emergency. Using SUTTON's (see GREEN and LANE[8]) atmospheric diffusion equations to calculate ground level concentrations at 5000 m assuming a 5 m/sec wind and adiabatic conditions, it can be shown that a release of just over 0.5 c of strontium-90 could result in an inhalation of 0.4 mμc by a person downwind and in the open air.

Alternatively the acceptable limit for inhalation may be calculated using the current ICRP concepts in which the environmental population are permitted a single intake of one-tenth of the amount permitted in 13-week continuous occupational intake. The $(MPC)_a$ for continuous exposure to strontium-90 is $10^{-10}\mu$ c/cm^3 for occupational exposure. Using the standard man breathing rate of 2×10^7 cm^3 per day the single intake for an adult in the environmental group is $2 \times 10^7 \times 10^{-10} \times \frac{1}{10} \times 91 = 18$ mμc. This is again a very low value.

As might be expected arguments for a maximum permissible inhalation based on the approaches outlined above are very restrictive and might result in an evacuation of the environmental population for several miles around a reactor site in the event of an emergency. The reasons why these restrictive values are obtained are that the MRC approach is meant to be applied to the dietary case where the intake might last for a period of days and the ICRP concept would allow similar intakes to be repeated at 13-week intervals and therefore is restrictive when applied to accidents on a "once in a lifetime" basis. In this paper the authors have made some calculations of maximum permissible inhalations based on the currently accepted maximum allowable bone dose and arrive at a less restrictive value.

PERMISSIBLE INHALATION BASED ON BODY BURDEN CONCEPT

The maximum permissible body burden (B) divided by the fraction (f_a) of the inhaled activity reaching the skeleton, gives a value for the maximum permissible inhalation of activity (I), i.e. $I = B/f_a$. The ICRP recommended value of the quantity B is 2 μc for strontium-90. There is some doubt about the appropriate value of f_a to use for strontium, from the ICRP recommendations $f_a = 0.12$ is given for strontium-90 and $f_a = 0.28$ for strontium-89. Experimental evidence from COMAR *et al.*[9] suggests that the strontium–calcium ratio in freshly deposited bone mineral is the same as that in blood for humans. On this evidence the same fraction of calcium and strontium is expected to be transferred to bone from the blood and this consideration could lead to an even higher value for f_a than either given above. It was therefore thought the more restrictive value of $f_a = 0.4$ which is that appropriate to the fraction of strontium transferred from air to blood should be used. Hence, $I = 2\ \mu c/0.4 = 5\ \mu c$. The value of B is for occupationally exposed workers, and one-tenth of this value may be taken for the special environmental group and hence the maximum permissible inhalation for a member of this group is 0.5 μc. Using the ICRP value for the effective half-life of strontium-90 in bone of 6400 days the mean dose to an adult skeleton in 50 years is 69 rems from an initial bone burden of 0.5 μc with no further intakes of activity. This may be compared with a mean dose of about 1500 rems to the bones which could be received by a worker continually exposed to the $(MPC)_a$ for strontium-89. In making this type of comparison it should be noted that the dose distribution in the bone will be different in each case.

In general the main objections to this kind of interpretation of ICRP recommendations are:

(a) The concept of maximum permissible body burden for bone seeking isotopes was derived from cases of chronic exposure to radium in which the distribution within the bone is somewhat different from the acute case and, for this and other reasons, may not strictly apply to acute exposure.

(b) In the case of young children who might

form part of the special group, the biological basis for the maximum permissible body burden is not so well founded because young children were not exposed to radium.

Notwithstanding the objections to this approach as given above it is thought that it serves as a useful guide to the magnitude of permissible inhalation.

PERMISSIBLE INHALATION BASED ON CALCULATIONS OF DOSE TO BONE

In this part of the paper, the results of experiments reported by various workers are used to calculate the dose to bone, and the maximum permissible inhalation is such that the dose due to strontium-90 is 1.5 rad per year, and 15 rads for strontium-89 following MRC recommendations. Two methods of calculation for strontium-90 have been used, and details are given in the following two sections. Strontium-89 and the presence of children in the environmental population are discussed later.

Section I

HARRISON *et al.*[10] have obtained data on the excretion and retention of Sr^{85} after a single intravenous injection. The study was carried out on two normal adult human subjects and good agreement was found between the data obtained from each. The experimentally determined retention of Sr^{85} over a period of 200 days may be expressed mathematically by either of the following equations

$$\% \text{ retention} = 15.4 + 40.4 \exp(-0.693t/1.7) + 29.8 \exp(-0.693t/4.4) + 14.4 \exp(-0.693t/50) \quad (1)$$

$$\% \text{ retention} = 28 \exp(-0.693t/2.7) + 51t^{-0.215} \quad (2)$$

where t is the time in days after the injection, and $t > 1$ in equation (2).

Both these equations give approximately the same value when used to calculate the dose to the skeleton in the first year. However, equation (2) will obviously give lower values of dose for each succeeding year and is probably more reasonable for prediction of very long term doses.

Equations (1) and (2) have been used to calculate the mean dose to the whole skeleton from strontium-90 plus yttrium-90 assuming uniform distribution. Table 1 shows the accumulated dose in rads for a 1 μc intravenous injection after 13 weeks, 1 year and the annual dose for the sixth year.

For a first annual dose of 1.5 rad to the skeleton, assuming uniform distribution, the single intravenous injection would be roughly 3 μc, which corresponds to an inhaled activity of $3\ \mu c/0.4 = 7.5\ \mu c$ of strontium-90. If the somatic mutation theory is accepted then the biological hazard is proportional to the integral dose over the tissue at risk. Hence, if the mean skeletal dose is the criterion for measuring hazard then a value of 7.5 μc of strontium-90 for the permissible inhalation over a short period could be accepted.

Table 1. Accumulated mean dose to bone for an intravenous injection of 1 μc of strontium-90

Period	Dose (rads)	
	equation (1)	equation (2)
13 weeks	0.19	0.18
1 year	0.51	0.53
6th year	0.41	0.27

Alternatively, if the maximum local dose is the important criterion of hazard then the mean dose to the bone is inapplicable because it does not take into account the non-uniform distribution of the activity in the skeleton. No experimental values for this non-uniformity factor have been obtained for human bones following a single acute intake of radiostrontium. However, the terminal non-uniformity factor in human bones has been measured for chronic intakes of radium by HINDMARSH *et al.*[11] and ROWLAND and MARSHALL[12] (1959). From these radium measurements the non-uniformity factor for strontium-90 deposits can be calculated assuming that the strontium is distributed in the same way as radium. OWEN and VAUGHAN[13] have made measurements of the non-uniformity factor in rabbit tibia for a single acute intake of radiostrontium. The pattern of bone growth in a rabbit resembles that in man,[14] and it is not unreasonable to take into account this value of non-uniformity in considering humans. The results of these groups of workers are summarized in Table 2. In the second column the ratio of the "hot spot" concentration to the uniform concentration is given for radium in human bones. Both maximum and mean values are given. The third column contains the mean value of D/D_∞ calculated by HINDMARSH, i.e. the dose to a small volume of tissue within a deposit of radiostrontium divided by the dose to the same volume of tissue when surrounded by a deposit of the same concentration but of infinite size. The value calculated by HINDMARSH is assumed to be applicable to ROWLAND's measurements. The factor, f, given in the next column is a product of the

Table 2. Summary of values for the non-uniformity factor for strontium-90

Reference	Radium: hot spot concentration / average concentration $= \frac{c}{c_0}$		Strontium: D/D_∞	Strontium: $f = \frac{c}{c_0} \times \frac{D}{D_\infty}$	Strontium: Non-uniformity factor	
	Max.	Mean	Mean	Mean	Mean	Max.
HINDMARSH *et al.*[11]	16	7	0.12	0.84	1.8	3.1
ROWLAND and MARSHALL[12]	83	40	0.12 assume	4.8	5.8	—
OWEN and VAUGHAN[14]	—	—	—	—	4	7

previous two columns and is the uncorrected non-uniformity factor. The maximum and minimum non-uniformity factors are given in the last column and have been increased by unity over the values of f, following the arguments of HINDMARSH, to take into account the slight overlap of dose effects between adjacent deposits. In the last line of the table the results of OWEN and VAUGHAN's rabbit experiments are given. The maximum value of 7 was obtained from a rabbit aged 14 months at injection and the mean value of 4 was the average over rabbits aged between $1\frac{1}{2}$ and 30 months. No variation of non-uniformity factors with age of injection was found in this age range.

In Table 2 it should be noted that the ratio C/C_0 as measured by HINDMARSH and by ROWLAND differ by about a factor of 5 which indicates the order of variability in this sort of measurement. Assessing these data it seems not unreasonable for a non-uniformity factor of 7 to be taken. This is the maximum value obtained in the single injection of strontium-90 in rabbits and it is slightly more than the mean value of about 6 obtained by ROWLAND. ROWLAND's data includes some obtained from humans who were injected with radium compounds in a short treatment period and hence these cases approximate to a single intake. Taking a non-uniformity factor of 7, then the inhaled activity of 7.5 μc/7 = 1.1 μc of strontium-90 would result in a maximum dose to the bone of 1.5 rad per year at particular independent sites such as Haversian canals.

Section II

COMAR *et al.*[9] have shown that the strontium–calcium ratio in newly formed bone is approximately the same as that in the circulating blood. HARRISON *et al.*[10] show a graph of the results of measurements of activity in the serum of an adult human after a single injection of radiostrontium. These results may be roughly represented for the first 30 hr by the following two-term exponential expression for the concentration ratio of strontium activity per g of calcium in the blood:

$$C = 0.37 \exp(-0.45t) + 0.09 \exp(-0.022t) \quad \frac{\mu c/g \text{ of Ca in blood}}{\mu c \text{ ingested}} \qquad (3)$$

where t is the time in hours after injection. Hence bone mineral which is forming at the time of inhalation will have maximum strontium-90 deposited in it and the amount of strontium-90 in later deposits will fall off as the plasma concentration decreases.

In order to make dose calculations for the newly deposited bone sites some basic assumptions must be made. Guidance for these assumptions has been obtained from the work of MARSHALL *et al.*[15] on the microscopic metabolism of calcium in adult canine bones. It is assumed that resorption cavities in human bones are produced and redeposited in much the same way and at much the same rate as in canine bones. The assumed dimensions of a human secondary Haversian system are given in Fig. 1(a). The outer diameter of the cavity is 200 μ, and it is redeposited until a central canal of only 40 μ exists. The cavity length is taken to be 2 mm from the serial section measurements made by MARSHALL and from ROWLAND[16].

MARSHALL measured the rate of deposition of calcium in adult dogs and found that the cavity diameter decreased roughly exponentially with time and could be represented by the following equation

$$b = b_0 \exp(-\beta t) \qquad (4)$$

where b_0 is the initial cavity diameter and b is the diameter after t hr, as shown in Fig. 1(b). The

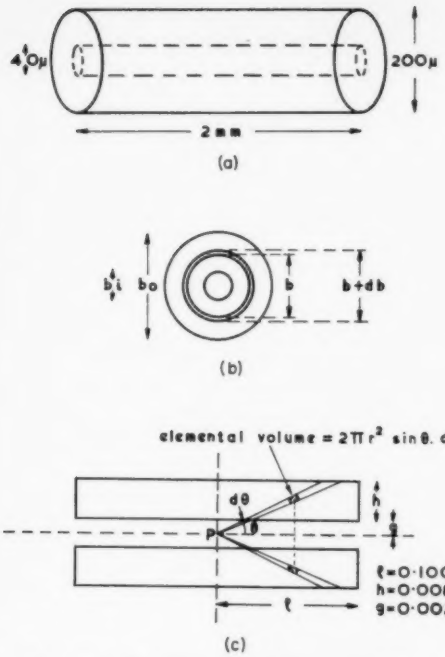

FIG. 1(a) Diagrammatic representation of a resorption cavity in a Haversian system at the beginning of deposition.

FIG. 1(b). Cross-section of the cavity.

FIG. 1(c) Diagram showing the polar co-ordinates used in calculating the dose at the centre of the cavity *P*.

constant $\beta = 0.001$ per hr, was experimentally determined for a number of dogs. From the knowledge of how the ratio strontium-90 per g of calcium in the blood varies with time after the initial intake and the time dependence of the calcium deposition in the hypothetical cavity, the total activity deposited can be calculated.

activity in an elemental

$$\text{volume of cavity} = c\rho \frac{\pi b}{2} db \text{ per unit length}$$

where c is the activity in μc of strontium-90 per g calcium and ρ is the weight of calcium (g) per cm^3 of bone.

Substituting the values of c and b from equations (3) and (4) and integrating over a long period of time, the following expression is obtained:

$$\text{total activity} = 4.6 \frac{b_0^2 \pi \beta}{2} \mu\text{c per unit length}$$

The mean activity per g of calcium per cm of activity is obtained by dividing by the total mass of calcium deposited per unit length, i.e.

$$\rho \frac{(b_0^2 - b_i^2)\pi}{4}$$

Thus mean activity

$$= 9.2\rho\ b_0^2 \pi\beta / \rho(b_0^2 - b_i^2)\pi \approx 9.2\beta$$
$$= 0.009\ \mu\text{c per g of Ca}$$

using a value of $\beta = 0.001$ per hr and neglecting b_i^2 relative to b_0^2.

The injection produced an initial concentration of 0.46 μc per g calcium in the blood which resulted in a mean activity of the deposited bone in the cavity of only 0.009 μc per g of calcium. In other words the single intake may be fifty times the constant intake level for the same total activity to be deposited in a cavity. In cavities as small as the one considered here the dose in the centre of the cavity is roughly proportional to the total activity in the cavity and almost independent of the distribution of the activity. It should be noted that in the above arguments the worst case is considered where the activity is deposited at the assumed maximum diameter.

The non-uniformity factor for an Haversian canal which has an active cavity with the dimensions given in Fig. 1(a) can readily be calculated. The dose (D) at point P, at the centre of a uniformly active deposit in an Haversian canal, may be written as

$$D = 2\pi\Delta \int_\theta \int_r \sin\theta\ .\ dr\ d\theta$$

The dose is expressed as a product of the number of β-particles per unit area at P and the mean rate of energy loss (Δ) of the particles. The integrations are carried out over the volume of the active deposit as indicated in Fig. 1(c) and n is the number of β-particles, per sec per cm^3 of the active bone. In this calculation the following assumptions are made:

(a) no scattering of the β-particles;
(b) the stopping power of soft tissue and bone is the same;
(c) the range of the β-particles from $Sr^{90} + Y^{90}$ is large compared with dimensions of the cavity. In fact less than 20 per cent of the total β-energy is associated with particles having a range shorter than 1 mm in soft tissue.

Integrating over the whole cylinder of active

deposit and using symbols given in Fig. 1(c) we obtain

$$D = 2n\Delta h\,\frac{\pi}{2} - \left(1 + \frac{g}{h}\right)\tan^{-1}\frac{(g+h)}{l} + \frac{g}{h}\tan^{-1}\left(\frac{g}{l}\right) + \frac{1}{h}\ln\sqrt{\left[\frac{(g+h)^2 + l^2}{g^2 + l^2}\right]} \quad (5)$$

Substituting the numerical values as shown in Fig. 1(c)

$$D = 2.9\ n\Delta h$$

The dose rate D_∞ in the centre of a small cavity surrounded by an infinite thickness of bone of the same specific activity n, is $D_\infty = n\bar{E}$, where $\bar{E}$ is the mean energy of the β-particles

hence $$\frac{D}{D_\infty} = \frac{2.9n\Delta h}{n\bar{E}} = \frac{2.9\Delta h}{\bar{E}}$$

Assume that the mean rate of energy loss $\Delta = \bar{E}/\bar{R}$ where $\bar{R}$ is the mean range which has a value of 0.5 cm in bone corresponding to $\bar{E} = 1.1$ MeV then

$$\frac{D}{D_\infty} = \frac{2.9h}{R} = 2.9\,\frac{0.008}{0.5} \simeq \frac{1}{20}$$

Hence this calculation shows that the dose at the centre of an active cylindrical deposit is twenty times less than the dose to the same point if it were surrounded by an infinite amount of active bone. The dose to points away from the centre of the deposit will clearly be less than that at the centre and so the calculation takes the worst possible case. It can also be shown that the dose due to active deposits of strontium on plane surfaces of non-active bone is less than for the canal-type deposits considered here.

The dose at the centre of any one active cavity will be increased by additional doses from neighbouring cavities. The mean distance between cavities may be roughly calculated for human bone from the work reported by ROWLAND[16] in which he examined a volume of about 0.12 cm^3 of bone taken from the tibia of a radium dial painter. This dial painter was exposed for a 2-month period at the age of 15 years and he died of osteogenic sarcoma at the age of 47 years. In this volume of bone seventeen hotspots were found hence the mean distance between cavities is about 2 mm. The dose from a point source of strontium-90 plus yttrium-90 in bone falls below 20 per cent of the maximum at a distance of 2 mm in bone.[17] Hence if a cavity is surrounded by six other cavities, each at a mean distance of 2 mm, then the dose at the centre of the cavity will be about double that due to its own activity. Therefore the value of D/D_∞ must be doubled to allow for this effect.

In comparing the effect of a single intake of strontium-90 with a continuous intake, two factors have to be considered. First, the resorption cavities which are being redeposited at the time of the single intake will lay down maximum concentrations of strontium almost immediately and the concentration will fall off rapidly as the plasma concentration drops. This changing concentration of strontium-90 means that the average activity per g of calcium is fifty times less than the initial concentration. The second factor allows for the fact that the dose in the centre of a small deposit of strontium is ten times less than it would be if it were surrounded by an infinite thickness of active bone. The same arguments could be used to produce similar but higher factors for the case of endosteal, periosteal and trabecular deposits of strontium-90 in bone mineral. These two factors reinforce one another and allow a single intake to raise the strontium-90/calcium ratio in the blood to 500 times (10×50) the constant level for continuous intake, for the same dose to the centre of an Haversian system. As calculated at the beginning of this paper, 0.15 mμc of strontium-90 in the 300 mg of calcium in the blood which corresponds to a bone deposition of 0.5 mμc per g of calcium, is taken to be the permissible continuous level of activity in the blood. Hence for a single intake the blood level may be raised to 0.15 mμc $\times$ 500 = 0.075 μc, i.e. an intake by inhalation of 0.075 μc/0.4 = 0.2 μc, and in this case the dose to the centre of a Haversian canal would not exceed 1.5 rad/year.

A more sophisticated method of calculation is to eliminate the parameter, t, between equations (3) and (4), in order to obtain the variation of the strontium-90 activity per g of calcium with diameter of the Haversian canal. This relationship is plotted in Fig. 2 using the numerical values already given in this paper. The noticeable feature of the deposition of the activity is that it forms an extremely narrow shell around the Haversian canal, a feature which has been noted in animal experiments. In principle, a precise experimental investigation of the shape of this curve together with the appropriate measurements of blood activity after a strontium-90 injection, should give information about the rate of deposition of calcium in the Haversian system.

DISCUSSION

The acceptable limits for inhalation of strontium-90 calculated in this paper are summarized in Table 3 where the results of similar calculations for strontium-89 are also given. In the first line of Table 3 the value of 1.0 μc is the amount of strontium-89, which, if inhaled,

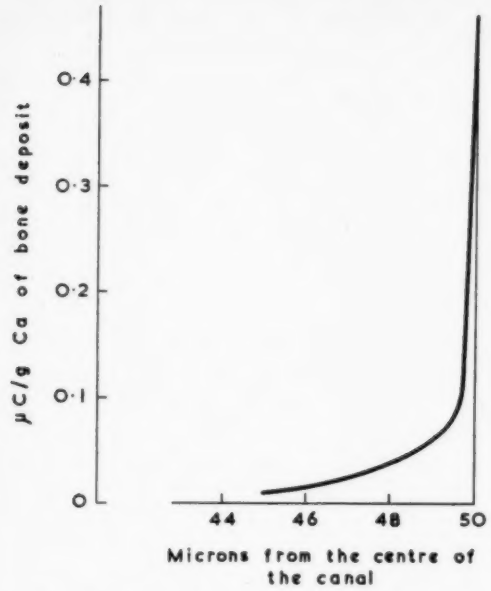

FIG. 2. Distribution of the activity of radiostrontium in a redepositing cavity in an Haversian canal. Initial diameter of the canal is taken to be 100 micron.

would give rise to one-tenth of the maximum permissible body burden immediately, but obviously this value dose not take into account the fairly rapid elimination and decay of the activity. Hence, although this method of calculation of an acceptable limit is not altogether unreasonable for isotopes with long biological half-lives it is too restrictive when applied to those with short half-lives. The values given in the second line are high because they are the result of a calculation of the mean dose to bone. This high value might be applicable if the somatic mutation theory of carcinogenesis is accepted in which the total dose is more important than the dose distribution. However, the mean dose may give a low assessment of the biological risk, and in the third line this mean has been reduced by applying a non-uniformity factor of 7 for strontium-90 and 16 for strontium-89. The higher value of 16 for strontium-89 was derived from theoretical considerations based on the β-particle energies and is relative to the experimental value of 7 for strontium-90. The final line of Table 3 gives the results of calculations of the dose to the centre of an Haversian canal on the basis of a permissible dose of 1.5 rad/year and 15 rad for strontium-90 and strontium-89, respectively.

From the values given in Table 3 it appears that a reasonable permissible limit of inhalation of strontium-90 by environmental populations in an emergency would be about 0.5 μc. This value of 0.5 μc is representative of the value derived from the body burden concept based on the human data for the radium body burden and the value derived from calculations summarized in the last two lines in the table. This

Table 3. Permissible limits for inhalation in an emergency

Sr^{90} (μc)	Sr^{89} (μc)	Remarks
0.5	1.0	Calculation of acute inhalation to produce a tenth of the recommended maximum permissible body burden
7.5	500	Mean values obtained from equations (1) and (2), assuming 1.5 rad/year and 15 rad for Sr^{90} and Sr^{89}, respectively
1.1	30	Values obtained from the line above by dividing by the non-uniformity factor
0.2	8	Calculation of the dose to the centre of an Haversian canal

is equivalent to an injection into the blood of 0.003 μc per kg for a standard man and, say, 0.04 μc per kg for a 5-kg baby. These values are about 15,000 and 1000 times, respectively, below the value of 44 μc per kg which is about the lower limit for observation of sarcoma in the bones of mice, as reported by FINKEL *et al.*[18]. The corresponding value for strontium-89, from Table 3 is about 16 μc. This value is only about thirty times greater than that for strontium-90 rather than 100 times greater, as given in the MRC Committee recommendations for dietary contamination. The reason is that the non-uniformity factor for strontium-89 is higher than for strontium-90 because the range of the β-particles from strontium-89 is less.

It is thought that a detailed discussion of the problems due to the presence of children in the environmental population would be out of place in this paper. In general children have been taken into account by using dose levels in the calculations which are a tenth of those recommended for occupationally exposed persons. The pertinent differences between adults and children in these considerations are breathing rate, skeleton weight and the amount of calcium in the plasma and body fluids. In considering the total dose to the skeleton, the ratio of the breathing rate to skeleton weight is important. In considering the activity per g of calcium deposited in Haversian systems the ratio of breathing rate to calcium is important. The authors consider that these two ratios in children can never be greater than perhaps three times similar ratios for adults. It has also been suggested that the tissues and bones of young children may be more radiosensitive than those of adults. The factor of 10 below occupational levels used in these calculations should allow a margin of safety for these differences between adults and children.

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SECRETION OF CALCIUM AND STRONTIUM INTO MILK*

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Abstract—The comparative secretion of Ca^{45} and Sr^{89} into the milk of goats and dairy cows under typical farm management was studied. The effects of calcium and stable strontium levels were investigated by using four diets containing 0.25 per cent Ca, 0.50 per cent Ca, 1.6–1.7 per cent Ca, and 0.25 per cent Ca plus 2.8–2.9 per cent stable strontium.

The comparative behavior of calcium and strontium was the same on all diets except for a possible renal effect with the goats. The Sr^{89}/Ca^{45} of the milk was consistently about 0.12 that of the dietary intake ($OR_{milk\text{-}diet} = 0.12$). The relative contributions to the discrimination against strontium have been estimated for the important physiological processes, i.e. absorption from the digestive tract, passage from blood to milk and renal excretion. On the medium-calcium diet at steady state the dairy cows secreted into milk about 0.08 per cent of the ingested Sr^{89} per liter; the goats, primarily because of body size, secreted much more, about 1.4 per cent of ingested Sr^{89} per liter of milk.

Since the $OR_{milk\text{-}diet}$ value did not vary with changes in dietary calcium levels it would be expected that long-term feeding of increased dietary levels of non-contaminated calcium would produce proportional decreases in the amounts of ingested radiostrontium appearing in milk; there would seem to be no advantage in the use of stable strontium for this purpose. Practical aspects are discussed.

THE contamination of the food chain by radiostrontium from nuclear detonations is well recognized and documented,[1–4] and the progression of radiostrontium along the food chain is at least qualitatively understood. The secretion of ingested radiostrontium into the milk of the dairy cow or goat is an important process for which reliable quantitative data should be obtained to permit evaluations and predictions of food contamination and to serve as a basis for action in the event of future contamination. Previous studies have given information that defines the gross behavior.[5–11] The present study was undertaken to obtain values under conditions of daily ingestion with dairy cows and goats under typical management. Special attention was given to the effects of alteration in dietary calcium and stable strontium levels. In addition to determination of comparative strontium and calcium behavior, values are also given for amounts of ingested radionuclides present in milk, tissues and excretions.

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METHODS

Procedures

In these studies, eight mature lactating goats of mixed breed and eight mature lactating Holstein cows were used. The cows were procured locally from tuberculosis-free herds and were shown by clinical examination to be free from brucellosis and mastitis. The animals were initially placed on experimental diets for 2 weeks under usual conditions, and then placed in metabolism stalls for an additional week before radionuclide administration (see Table 1 for summary of experimental treatment). After the animals were accustomed to the stalls, they were given Ca^{45} and Sr^{89} twice daily by capsule for 9 or 10 days. The goats received about 0.2 mc of Ca^{45} and 0.1 mc of Sr^{89} and the cows received about 1 mc of Ca^{45} and 0.7 mc of Sr^{89} per capsule. The milk, urine and feces were quantitatively collected and aliquots assayed for Ca^{45} and Sr^{89} by the double

Table 1. *Protocol of animals and diets*

Goat no.	Body wt. (lb)	Average milk yield (l/day)	Calcium in diet (%)	Strontium in diets (%)	Average daily intake of calcium and strontium (g)
3	79	0.58	0.25	—†	4.9
6	88	0.92	0.25	—	4.9
7	59	0.47	0.50	—	10.6
10	70	0.33	0.50	—	10.6
4	78	0.43	1.7	—	36.0
5	87	0.37	1.7	—	36.0
8	68	0.60	0.25	3.0	105.8
9	66	0.46	0.25	3.0	105.8
Cow no.					
3	1068	16	0.25	—	45
10	1036	10	0.25	—	45
5	1210	11	0.50	—	90
8	1132	12	0.50	—	90
4	912	13	1.6	—	281
6	1180	7.4	1.6	—	281
7	920	8.2	0.25	2.8	693
9	1076	8.8	0.25	2.8	693

† Indicates no stable strontium added to diet.

counting procedure as described by COMAR.[12] Blood was taken by jugular puncture twice daily and the radionuclide content of plasma determined. The radionuclide data are expressed as "percentage of the daily ingested dose".

The diet consisted of timothy hay (about 0.3 per cent Ca) and a grain mixture supplemented with varying levels of calcium or strontium, added either as $CaHPO_4$ or $SrHPO_4$. The basal grain diet was as follows: wheat bran, 13.1 per cent; standard middlings, 8.1 per cent; Brewer's grains, 10.1 per cent; corn dried distillers grain, 27.3 per cent; soybean oil meal, 8.1 per cent; hominy, 11.1 per cent; ground oats, 7.1 per cent; salt, 1.0 per cent; rice bran, 3.0 per cent; cane molasses, 11.1 per cent. The unsupplemented basal grain ration contained about 0.17 per cent Ca and was used as the low-calcium grain mixture. Sufficient $CaHPO_4$ was added to the basal diet to yield a grain mixture containing about 0.77 per cent Ca (normal) and 3.9 per cent Ca (high). The calcium to phosphorus ratios of the low-, normal- and high-calcium diets were found to be about 0.78, 0.93 and 1.15, respectively. The high-strontium grain mixture contained about 6.3 per cent Sr and 0.17 per cent Ca. The goats consumed about 1.2 kg of hay and 0.9 kg of grain mixture per day; the cows, on the average, consumed about 10.4 kg of hay and 7.3 kg of grain. The percentage of calcium in the total diet of the goats was approximately 0.25 per cent, 0.50 per cent and 1.7 per cent Ca for the low-, normal- and high-calcium diet and 0.25 per cent Ca and 2.8 per cent Sr for the high-strontium diet. The percentage of calcium in the total ingested diet for the cows was the same as for the goats except that the high-calcium diet contained 1.6 per cent Ca, and high-strontium 3.0 per cent Sr and 0.25 per cent Ca.

At the end of the collection period, the animals were killed and tissues taken for radioassay by standard procedures.

Expression of results

To characterize the differential behavior in a metabolic system of two elements such as strontium and calcium COMAR *et al.*[13] proposed the term "strontium–calcium observed ratio" (*OR*) to denote the overall differential behavior in the accumulation of the two elements in tissue or sample from its precursor as shown by the relative concentrations at steady state. To denote the discrimination that is produced by a given physiological process, the term "strontium–calcium discrimination factor" (*DF*) was defined. It is emphasized that the discrimination factor, as defined, takes into account not only the

relative discrimination that occurs in a given process, but also the net effect on the Sr/Ca ratio in the tissue or sample as governed by the absolute amounts retained. Since this matter has caused some confusion, perhaps a simple illustration will serve for clarification. Assume that an animal has in its body at a given time 100 units each of strontium and calcium. If it excretes in the urine 10 units of Sr and 1 unit of Ca then the body burden would be 90 units of Sr and 99 units of Ca or a Sr/Ca ratio of 90/99. If the animal were instead to excrete 1 unit of Sr and 0.1 unit of Ca then the body burden would be 99 units of Sr and 99.9 units of Ca to give a Sr/Ca ratio of 99/99.9. In other words, the animal has in both instances excreted ten times more Sr than Ca, but the effect produced in the body is much greater in the first instance. It is clear then that the effect of any discriminatory process is dependent upon the amounts retained in the tissue or secretion in question. It is also pointed out that the amount retained is usually governed by several processes.

In a paper of WASSERMAN *et al.*[6] there was developed a series of equations to permit the calculation of *OR* and *DF* values from measurements following oral administration of radionuclides of strontium and calcium to a lactating animal. The following assumptions were made on the basis of previous experience: (a) the only processes important in causing discrimination in a lactating animal are absorption, urinary excretion and secretion into milk; (b) the urinary excretion of each element is proportional to the amount absorbed; and (c) the amount secreted into milk is proportional to the amount absorbed minus that excreted into the urine. The equations are as shown in Table at foot of page.

GARNER *et al.*[14] have extended this approach to take into account the possibility of a discrimination effect from endogenous fecal secretion. It should be pointed out that their results confirm the assumption of WASSERMAN *et al.*[6] that there is no significant discrimination via the endogenous fecal route for strontium and calcium; there was, however, an endogenous effect when comparisons were made

Comparative movement of strontium and calcium into tissues or excretions	$\text{Observed ratios, } OR_{\text{sample/precursor}} = \dfrac{\text{Sr/Ca of sample}}{\text{Sr/Ca of precursor}}$
Equation for contribution of physiological processes to overall discrimination between strontium and calcium	$OR = (DF_{\text{absorptive}})\ (DF_{\text{urinary}})\ (DF_{\text{lactational}})$
Absorptive discrimination	$DF_{\text{absorptive}} = \dfrac{100 - F_{Sr}}{100 - F_{Ca}}$
Urinary discrimination	$DF_{\text{urinary}} = \dfrac{(100 - F_{Ca}) - U_{Sr} \times \dfrac{100 - F_{Ca}}{100 - F_{Sr}}}{B_{Ca} + L_{Ca}}$
Lactational discrimination on body	$DF_{\text{lactational-body}} = \dfrac{B_{Sr}}{B_{Ca}} \times \dfrac{B_{Ca} + L_{Ca}}{B_{Sr} + L_{Sr}}$
Lactational discrimination on milk	$DF_{\text{lactational-milk}} = \dfrac{L_{Sr}}{L_{Ca}} \times \dfrac{B_{Ca} + L_{Ca}}{B_{Sr} + L_{Sr}}$

F_{Sr} and F_{Ca} = per centage of ingested dose of Sr and Ca, respectively, in the feces.
B_{Sr} and B_{Ca} = per centage of ingested dose of Sr and Ca, respectively, in the body.
L_{Sr} and L_{Ca} = per centage of ingested dose of Sr and Ca, respectively, in the milk.
U_{Sr} and U_{Ca} = per centage of ingested dose of Sr and Ca, respectively, excreted in the urine.

with barium. The equations of GARNER *et al.*[10] are reproduced below and it is noted: (a) that measurements are required after both oral and intravenous administration, and (b) that when there is no fecal endogenous discrimination the equations reduce to those as quoted above.[6]

$$OR_{\text{sample-precursor}} = \frac{Sr^{89}/Ca^{45} \text{ of sample}}{Sr^{89}/Ca^{45} \text{ of precursor}}$$

$$DF_{\text{absorptive}} = \frac{\% \text{ of orally administered } Sr^{89} \text{ absorbed}}{\% \text{ of orally administered } Ca^{45} \text{ absorbed}}$$

$$DF_{\text{urinary}} = \frac{100 - \% \text{ of intravenously administered } Sr^{89} \text{ excreted in urine}}{100 - \% \text{ of intravenously administered } Ca^{45} \text{ excreted in urine}}$$

$$DF_{\text{fecal}} = \frac{100 - \% \text{ of intravenously administered } Sr^{89} \text{ excreted in feces}}{100 - \% \text{ of intravenously administered } Ca^{45} \text{ excreted in feces}}$$

$$DF_{\text{lactational-milk}} = \frac{OR_{\text{milk-diet}}}{DF_{\text{absorptive}} \times DF_{\text{urinary}} \times DF_{\text{fecal}}}$$

$$OR_{\text{body-diet}} = DF_{\text{absorptive}} \times \frac{\% \text{ of intravenously administered } Sr^{89} \text{ retained in body}}{\% \text{ of intravenously administered } Ca^{45} \text{ retained in body}}$$

$$DF_{\text{lactational-body}} = \frac{OR_{\text{body-diet}}}{DF_{\text{absorptive}} \times DF_{\text{urinary}} \times DF_{\text{fecal}}}$$

Table 2. Individual milk, plasma and excretion values per goat no. 10
(body wt., 70 lb; 0.50 per cent calcium diet: received eighteen oral dosages each containing 0.185 mc Ca^{45} and 0.087 mc Sr^{89})

Days of dosage	Time of milking	Milk			Plasma		Urine			Feces		
		Amt. (l.)	Ca^{45} (% of daily dose/l.)	Sr^{89} (% of daily dose/l.)	Ca^{45} (% of daily dose/l.)	Sr^{89} (% of daily dose/l.)	Amt. (lb)	Ca^{45} (% daily dose/day)	Sr^{89} (% daily dose/day)	Amt. (lb)	Ca^{45} (% daily dose/day)	Sr^{89} (% daily dose/day)
1st	p.m.	0.14	1.3	0.17	0.27	0.053	0.81	0.60	0.42	3.5	36	34
2nd	a.m.	0.22	6.5	0.80	0.57	0.14	1.0	1.8	1.3	3.5	45	45
	p.m.	0.14	9.0	1.2	0.63	0.15						
3rd	a.m.	0.22	12	1.4	0.80	0.20	0.75	1.0	1.1	4.1	15	16
	p.m.	0.14	14	1.8	0.80	0.18						
4th	a.m.	0.22	14	1.7	0.81	0.25	1.1	1.0	1.7	3.4	74	73
	p.m.	0.14	14	2.1	0.77	0.19						
5th	a.m.	0.22	14	1.9	0.75	0.20	0.88	0.95	1.4	3.5	71	79
	p.m.	0.08	14	1.8	0.83	0.22						
6th	a.m.	0.25	13	1.7	0.79	0.20	0.81	1.2	1.5	3.5	55	57
	p.m.	0.14	13	1.7	0.72	0.22						
7th	a.m.	0.12	13	1.5	0.70	0.20	1.3	2.0	2.0	3.1	125	114
	p.m.	0.16	12	1.7	0.75	0.22						
8th	a.m.	0.19	12	1.7	0.78	0.20	1.1	1.9	2.0	2.9	81	82
	p.m.	0.11	14	1.9	0.75	0.21						
9th	a.m.	0.16	14	1.8	0.76	0.19	1.4	1.7	2.0	3.1	73	76
	p.m.	0.11	13	1.8	0.81	0.21						
10th	a.m.	0.22	13	1.7								

RESULTS

Table 2 presents values for the levels of Ca^{45} and Sr^{89} appearing in the individual milk, plasma, urine and feces collections from one of the goats during the 9-day experimental period. The pattern is representative and similar to that for all the other goats. It may first be noted that the plateau levels of the radionuclides in milk and plasma were reached by the third day after the start of the administration. For the feces and urine, it appeared that plateau levels were reached by the second day. Table 3 presents similar values for one of the cows; plateau levels in milk and plasma were reached by the fourth to sixth day and steady-state excretion values were essentially attained by the third day. The values in Tables 2 and 3 are presented mainly to show the time pattern and the order of agreement between individual samples. Discussion of the magnitude of the values is deferred until presentation of averages of all animals used.

Table 4 presents a summary of values for the goats. It is first noted that an $OR_{\text{milk-diet}}$ of 0.12 was obtained for all diets. Likewise there was no significant effect of diet on the $OR_{\text{plasma-diet}}$ (0.24–0.34), the $OR_{\text{milk-plasma}}$ (0.37–0.49), or the $OR_{\text{feces-diet}}$ of 1.2. There did seem to be an effect on the renal behavior with a greater comparative Sr^{89} excretion at the lower calcium level as shown by higher values of $OR_{\text{urine-diet}}$ and $OR_{\text{urine-plasma}}$.

Table 3. Individual milk, plasma and excretion values per cow no. 8
(Body wt. 1132 lb; 0.50 per cent calcium diet; received seventeen oral dosages each containing 0.91 mc Ca^{45} and 0.75 mc Sr^{89})

Days of dosage	Time of milking	Milk			Plasma			Urine			Feces		
		Amt. (l.)	Ca^{45} (% of daily dose/l.)	Sr^{89} (% of daily dose/l.)	Ca^{45} (% of daily dose/liter)	Sr^{89} (% of daily dose/liter)	Amt. (lb)	Ca^{45} (% daily dose/day)	Sr^{89} (% daily dose/day)	Amt. (lb)	Ca^{45} (% daily dose/day)	Sr^{89} (% daily dose/day)
1st	a.m.	9.1	0.066	0.024	0.019	0.0034	11	0.15	0.086	56	4.0	6.2
	p.m.	5.4	0.21	0.035	0.034	0.0055						
2nd	a.m.	8.6	0.41	0.035	0.056	0.0082	16	1.7	0.64	67	31	46
	p.m.	5.1	0.57	0.042	0.053	0.0079						
3rd	a.m.	8.8	0.66	0.062	0.066	0.0097	15	2.3	1.2	84	54	72
	p.m.	5.0	0.78	0.060	0.072	0.011						
4th	a.m.	8.5	0.76	0.065	0.086	0.012	14	1.8	1.2	79	52	74
	p.m.	5.0	0.79	0.071	0.069	0.013						
5th	a.m.	8.9	1.0	0.075	0.090	0.014	14	1.9	1.2	78	56	76
	p.m.	4.5	0.52	0.060	0.093	0.014						
6th	a.m.	7.5	1.2	0.087	0.10	0.015	17	1.8	1.4	71	64	90
	p.m.	4.7	1.2	0.096	0.11	0.016						
7th	a.m.	7.6	1.1	0.094	0.099	0.013	24	1.4	1.1	79	72	95
	p.m.	5.6	1.1	0.088	0.10	0.014						
8th	a.m.	9.2	0.48	0.064	0.086	0.012	37	1.1	1.2	92	78	100
	p.m.	4.2	1.1	0.083	0.082	0.012						
9th	a.m.	8.6	0.98	0.082	0.083	0.012	38	0.26	0.92	73	64	81

Table 4. Summary of comparative and absolute calcium and strontium behavior in goats

	Diet			
	Low-calcium (0.25%)	Medium-calcium (0.50%)	High-calcium (1.7%)	High-strontium (3.00% strontium, 0.25% calcium)
$OR_{milk-diet}$	0.12	0.12	0.12	0.12
$OR_{plasma-diet}$	0.24	0.25	0.31	0.34
$OR_{milk-plasma}$	0.49	0.48	0.42	0.37
$OR_{urine-diet}$	3.2	1.2	0.74	0.86
$OR_{urine-plasma}$	14	4.8	2.7	2.5
$OR_{feces-diet}$	1.2	1.2	1.2	1.2
$DF_{absorptive}$	0.34	0.30	0.29	0.31
$DF_{urinary}$	0.50	0.58	0.52	0.63
$DF_{lactational-milk}$	0.40	0.44	0.44	0.47
$DF_{lactational-body}$	1.2	1.1	1.1	1.0
Milk				
Ca^{45} (% daily dose/l.)	14	12	6.4	6.2
Sr^{89} (% daily dose/l.)	1.7	1.4	0.74	0.77
Ca^{45} (% daily dose/day)	11	4.6	2.4	3.2
Sr^{89} (% daily dose/day)	1.2	0.54	0.28	0.41
Feces				
Ca^{45} (% daily dose/day)	62	78	86	80
Sr^{89} (% daily dose/day)	87	94	96	94
Urine				
Ca^{45} (% daily dose/day)	1.4	1.6	2.2	3.0
Sr^{89} (% daily dose/day)	4.6	1.9	1.6	1.6

There seemed to be no significant effect of diet on the various discrimination factors (*DF* values); the effect of the altered urinary discrimination was less marked when expressed as the *DF* value, which expresses the contribution of this physiological process to the overall discrimination.

The amount of ingested radionuclides that reached the milk when expressed as percentage of daily dose per liter varied from about 13 per cent Ca^{45} and 1.5 per cent Sr^{89} at medium- and low-calcium to about 6.3 per cent Ca^{45} and 0.75 per cent Sr^{89} at high-calcium and high-strontium levels. When expressed as percentage of daily dose per day it was of interest to note that there was more nearly an inverse proportional relationship between level of dietary alkaline earths and amounts of ingested radionuclide of calcium and strontium secreted into the milk. It is especially important to note that the increased dietary strontium had about the same effect on the Ca^{45} and Sr^{89} as did the increased dietary calcium.

The fecal excretion of Ca^{45} and Sr^{89} did indicate a possible increased absorption at the low-calcium level, but the values are difficult to interpret because small errors in collection and analysis became magnified in the subtraction of two large numbers to give an absorption value (amount ingested — amount in feces). There were no large differences in the urinary excretion values as a result of diet; it is difficult to assign any meaning to such changes as were noted because the urinary values reflect both changes in amount absorbed and alterations in the renal function.

Table 5. Summary of comparative and absolute calcium and strontium behavior in cows

	Diet			
	Low-calcium (0.25%)	Medium-calcium (0.50%)	High-calcium (1.70%)	High-strontium (2.8% strontium, 0.25% calcium)
$OR_{milk-diet}$	0.10	0.092	0.11	0.12
$OR_{plasma-diet}$	0.18	0.18	0.13	0.21
$OR_{milk-plasma}$	0.58	0.52	0.80	0.59
$OR_{urine-diet}$	1.4	1.1	3.2	0.66
$OR_{urine-plasma}$	8.4	6.4	7.3	3.2
$OR_{feces-diet}$	1.4	1.2	1.2	1.2
$DF_{absorptive}$	0.22	0.14	0.34	0.24
$DF_{urinary}$	0.84	0.66	0.73	0.68
$DF_{lactational-milk}$	0.58	0.96	0.42	0.76
$DF_{lactational-body}$	1.4	0.97	1.5	1.2
Milk				
Ca^{45} (% daily dose/l.)	1.2	0.82	0.40	1.0
Sr^{89} (% daily dose/l.)	0.14	0.080	0.041	0.12
Ca^{45} (% daily dose/day)	14	10	4.2	8.1
Sr^{89} (% daily dose/day)	1.4	0.90	0.38	0.98
Feces				
Ca^{45} (% daily dose/day)	69	73	90	79
Sr^{89} (% daily dose/day)	93	96	97	95
Urine				
Ca^{45} (% daily dose/day)	0.51	1.2	1.7	2.7
Sr^{89} (% daily dose/day)	1.4	1.3	1.2	2.0

Table 5 presents parallel summary values for the dairy cows. In regard to both observed ratios and discrimination factors, it is noted that there was no significant effect of the calcium and strontium levels in the diet. In contrast to the goats, no clear effects on urinary discrimination were observed. Generally, an increased dietary calcium decreased the amounts of radionuclides entering the milk although there was no strict proportionality; the reasons for this will be discussed in a later section. It also appeared as if the high stable strontium had less of an effect on the amount of radionuclides appearing in the milk than did the high level of dietary calcium. The fecal and urinary excretions of Ca^{45} and Sr^{89} were similar to the values for the goats.

Table 6 presents values for the comparative deposition of calcium and strontium in various tissues. The values are given in terms of observed ratios. There appeared to be no striking effects of diet, although more animals would be required for a definitive determination on this point. Values from all animals are averaged for convenience of comparison among tissues. For the goats it appeared that all tissues had about the same *OR* value as the plasma (0.28), except for the teeth, with a higher value of 0.65; this is in agreement with earlier findings.[6] For the cows also, the teeth appeared to have significantly higher values than the other tissues. The definitely high *OR* values for the teeth can undoubtedly be explained by surface adsorption of the alkaline earths during ingestion and rumination. There seemed to be a greater range of values among the tissues of

Table 6. Summary of comparative deposition of calcium and strontium in tissues
(Values expressed as $OR_{\text{tissue-diet}}$ and represent mean of two animals; average represents all diets and standard error of mean)

Tissue	Goat					Cow				
	Low-Ca	Medium-Ca	High-Ca	High-Sr	Average	Low-Ca	Medium-Ca	High-Ca	High-Sr	Average
Rib	0.28	0.27	0.23	0.30	0.27 ± 0.02	0.24	0.16	—	0.22	0.21 ± 0.03
Tibia	0.29	0.28	0.23	0.34	0.28 ± 0.02	0.22	—	—	0.32	0.27 ± 0.08
Vertebra	0.26	0.26	0.22	0.30	0.26 ± 0.01	0.22	0.16	—	0.15	0.18 ± 0.03
Mandible	0.28	0.26	0.22	0.27	0.26 ± 0.02	0.24	0.18	—	0.14	0.19 ± 0.04
Tooth	0.60	0.45	0.56	1.0	0.65 ± 0.12	0.48	—	—	—	0.48 ± 0.11
Kidney	0.32	0.38	0.27	0.41	0.34 ± 0.02	0.37	0.28	—	0.34	0.33 ± 0.05
Liver	0.22	0.25	0.20	0.30	0.24 ± 0.02	0.42	0.25	—	0.16	0.29 ± 0.07
Spleen	0.28	0.24	0.30	0.28	0.28 ± 0.02	0.25	0.16	—	0.16	0.19 ± 0.03
Muscle	0.20	0.21	0.20	0.30	0.23 ± 0.02	0.21	0.12	—	0.20	0.18 ± 0.02
Plasma	0.24	0.25	0.31	0.34	0.28	0.18	0.18	—	0.21	0.17

the cows with some suggestion of a higher kidney value.

DISCUSSION

Comparative secretion of Sr *and* Ca *into milk*

The comparative secretion of strontium and calcium from the diet into milk is of primary importance in connection with relationships between environmental contamination and the body burden of Sr^{90} developed in the human population. In 1958, WASSERMAN *et al.*[6] summarized the consensus of observations at that time and pointed out that in cows and goats the Sr^{90}-to-Ca ratio of milk ranged from 0.09 to 0.16 that of the ration being consumed by the animal (i.e. $OR_{\text{milk-diet}} = 0.09$ to 0.16). Observations since then in human beings[15] and domestic animals under varying management[7,10,11] have confirmed this finding; identical results are obtained whether one uses: (a) stable calcium and strontium values in feed and milk; (b) stable calcium and fallout Sr^{90} values; or (c) experimentally given radionuclides of strontium and calcium. The results in this paper serve to strengthen the observation since the average $OR_{\text{milk-diet}}$ values for eight cows and eight goats were 0.11 and 0.12, respectively.

Although it is emphasized that the $OR_{\text{milk-diet}}$ cannot be considered as a biological constant, it is noted that the values appear to fall within a relatively narrow range for all situations so far reported. The concept of the *OR* value can then be used to advantage for prediction purposes, always with the limitations kept in mind as to the applicability of the particular *OR* value employed. It is useful to estimate the total dietary intake of Sr^{90} and calcium of dairy cattle by calculation from values in milk. There seems little question but that dividing the Sr^{90}/Ca measured in milk by 0.1 (typical value for $OR_{\text{milk-diet}}$) would give an estimate of dietary intake more simply and more accurately than would efforts to measure total intakes directly or to sum the Sr^{90} contributions of components of the feed and water. Also, an estimate of the Sr^{90}/Ca being laid down in the newly forming and exchangeable bone of the animals could be gained by dividing the Sr^{90}/Ca in milk by 0.5 (typical value for $OR_{\text{milk-plasma}}$).

It is next of interest to consider the amounts of ingested radiostrontium and radiocalcium that reach the milk. VISEK *et al.*[16] showed that the amount of injected Ca^{45} that appeared in milk was directly related to the amount of milk secreted; it would therefore be expected that the Sr^{90} would behave in the same way because of the relatively constant $OR_{\text{milk-diet}}$ values. GARNER and SANSOM[10] and EKMAN and ÅBERG[9] have presented data that support this view. Therefore it appears more useful to discuss amounts in terms of percentage of

ingested dose per liter of milk rather than per total amount of milk secreted.

COMAR and WASSERMAN[5] in 1956 reported that two cows on a daily calcium intake of 25 g averaged 0.20 per cent of the daily dose of radiostrontium per liter milk whereas the same cows on an intake of 100 g calcium per day gave a value of 0.13 per cent. As can be seen from Table 5, cows on what is regarded as a normal calcium level (0.50 per cent) secreted about 0.08 per cent of the ingested radiostrontium per liter. GARNER and SANSOM[10] have also reported a value of 0.08 per cent and it is considered that this value is more reliable for typical management conditions; CRAGLE and DEMOTT[7] have reported a value of 0.07 per cent. This value would be expected to be higher at lower calcium intakes and conversely lower at higher calcium intakes.

From Table 4, it may be noted that goats on the medium calcium level (0.5 per cent) secreted about 1.4 per cent of the ingested radiostrontium per liter of milk. WASSERMAN *et al.*[6] reported values of 0.61 and 1.3 per cent for two goats. These are somewhat lower than the values of from 3.1 to 3.8 per cent calculated from the data of EKMAN and ÅBERG[9], but it must be remembered that there is considerable dependency on the dietary calcium intake.

It is of interest to note that the percentage of ingested radiostrontium secreted per liter of milk is more than ten times higher for the goat than for the cow. This comes about because the goat puts a much larger proportion of its dietary calcium into the milk per liter than does the cow. This differential behavior is of little practical significance from the standpoint of environmental contamination because the $OR_{\text{milk-diet}}$ values are the same for the two species and if the cow and goat consume the same feeding stuffs (i.e. feeds with same Sr^{90}/Ca ratio), about the same Sr^{90}/Ca ratio will be in the milk. As will be discussed later (Table 7), the cow tends to put more of its skeletal calcium into milk than does the goat; thus there may be situations following short-term increases of dietary Sr^{90} in which cow's milk would have a lower Sr^{90}/Ca ratio than goat's milk. This would be expected especially if the cows were on sub-optimal calcium intake. If the cow and goat consume the same amount of radiostrontium, then the concentration in the goat's milk will be tenfold higher; this observation may be useful for anyone interested in the production of Sr*-containing milk.

In regard to the amount of ingested calcium appearing in milk, it is important to recall that the calcium content of cow's milk tends to remain reasonably constant at about 1.1 g of calcium per liter regardless of dietary intake. This means that a cow ingesting 45 g of calcium per day should secrete Ca equal to 2.4 per cent of the ingested calcium per liter of milk, one ingesting 90 g should secrete about 1.2 per cent, and one ingesting 281 g should secrete about 0.4 per cent. From Table 5 it can be noted that the actual amounts of Ca^{45} secreted at these levels of intake were 1.2, 0.82 and 0.4 per cent, respectively. The extent to which the Ca^{45} is lower than the stable calcium value is a measure of the contribution of skeletal calcium to the milk and such contributions can be calculated by the usual procedures of isotope dilution. These data are presented in Table 7. It is of considerable interest to note that about 50 per cent of the milk calcium was coming from the skeleton of the cows on the low and medium diet and from the goats on the low-calcium diet. In a study of calcium secretion in human milk this same behavior was noted.[15] Aside from nutritional implications, this matter should be taken into account in the interpretation of Sr^{90}–Ca survey values in milk especially if

Table 7. Origin of milk calcium

Ca in diet (%)	Daily calcium intake (g)	Origin of calcium	
		From diet (%)	From skeleton (%)
	Cows		
0.25	45	39	60
0.50	90	50	50
1.6	281	98	2
	Goats		
0.25	4.9	49	51
0.50	10.6	90	10
1.7	36.0	100	—

intake levels of Sr^{90} were to be changing fairly rapidly.

The discrimination factors (*DF* values) are calculated to permit an estimation of the contribution of given physiological processes to the overall differential behavior of the two elements. The *DF* values for the goats (Table 4) are in general agreement with those reported previously by WASSERMAN *et al.*[6] The *DF* values for the cows (Table 5) are in good agreement generally with those reported by GARNER and SANSOM[10].

It may be first noted from Table 4 (goats) that there was no observable effect of dietary calcium and strontium level on the *DF* values. It is clear that absorptive discrimination, that is, preferential absorption of calcium from the gastrointestinal tract, was of major importance in bringing about the differential secretion into milk ($DF_{\text{absorptive}} = 0.29–0.34$). Lactational discrimination ($DF_{\text{lactational-milk}} = 0.40–0.47$) was next in importance. Urinary discrimination was equally significant ($DF_{\text{urinary}} = 0.50–0.63$) even though the total urinary excretion of the alkaline earths was small. Urinary excretion and also the secretion from blood to milk make an important contribution to overall discrimination because of the relatively small amounts of the two elements that are absorbed and metabolized by the body. The discrimination that occurs in the lactation process of course tends to raise the $OR_{\text{body-diet}}$ value ($DF_{\text{lactational-body}} = 1.0–1.2$). As can be noted from the *DF* values this effect is small; theoretically the $DF_{\text{lactational-body}}$ should be greater than 1 when the $OR_{\text{milk-plasma}}$ or the $DF_{\text{lactational-milk}}$ are less than 1, and any values of 1 or less reflect experimental error.

The *DF* values for the cows (Table 5) show greater dispersion than for the goats, but here also there seemed to be no consistent effect of dietary level of calcium or strontium. Absorptive discrimination was even more important than it had been for the goats ($DF_{\text{absorptive}} = 0.14–0.34$). The urinary discrimination, conversely, was much less important and this was because of the low urinary excretion of both calcium and strontium by the cow.

Of particular interest is the question as to the extent that increased dietary calcium or strontium will decrease the amount of ingested radiostrontium that appears in the milk. The pros and cons of this matter have been discussed at length in the literature[17–20] and it is apparent that the matter is not a simple one. The consensus is that increased dietary calcium in a physiologically normal diet does tend to reduce proportionally the body burden of ingested radiostrontium. If it can be assumed that the body burden of radiostrontium is lowered, it would be expected that the level in the body fluids would be lowered similarly and, as a consequence, one would expect a similar decrease in the amount secreted into milk. It must be remembered that the present studies are essentially short-term in two respects: the animals had been on a modified diet for 3 weeks and therefore any adaptation could not have been completed; also the double tracer method was carried out over a 9- to 10-day period.

In terms of the effect of dietary calcium on amounts of ingested radiostrontium entering the milk it was noted in goats that doubling the calcium (from 0.25 to 0.5 per cent) decreased the Sr^{89} only from 1.7 to 1.4 per cent of the daily dose per liter; in cows there seemed to be more of an effect with a decrease of almost twofold from 0.14 to 0.08 per cent. When the calcium was increased almost threefold from 0.5 per cent to 1.6–1.7 per cent the Sr^{89} in both goat's and cow's milk decreased about twofold.

It is emphasized, however, that in this type of study little meaning can be attached to the absolute values of Sr^{89} appearing in milk. This is because the Ca^{45}, which follows the stable calcium, did not decrease in direct proportion to the increased dietary level; the reasons for such behavior have been discussed previously.[2,20] Actually there was no effect on the $OR_{\text{milk-diet}}$ values and this suggests that the long-term feeding of increased dietary levels of calcium to lactating animals would be expected to result in a proportional decrease of ingested radiostrontium in milk. The observations also suggest that there is no advantage to be gained from the use of stable strontium instead of stable calcium for this purpose. The definite answer to this question must come from long-term feeding studies in which animals are raised

for periods of a year or more on differing levels of dietary calcium. Such studies are in progress in this laboratory.

Modification of dietary calcium levels

A full discussion of remedial measures for the reduction of Sr^{90} levels in food is not within the scope of this paper; however, it does seem pertinent to present some viewpoints in relation to modifications of dietary calcium levels. Clearly, any undertaking of remedial measures should result only from counsel of responsible authorities since such decisions must be based on medical assessment of probable harm from Sr^{90} weighed against all other factors such as efficiency of the procedure and possible harm from the procedure. Also there must be taken into account any overall program of remedial measures. If, for example, procedures were being used to decontaminate milk by ion exchange procedures, there would be little usefulness in trying to reduce the Sr^{90} content by factors of about 2 through dietary means. On the other hand, if a series of remedial measures were being employed, then a reduction factor of about 2 anywhere along the chain might be multiplied in end effect and thereby be useful. Decisions are most difficult at present because we have little information on the relationships between possible effects and dietary intakes at present environmental levels.

It is clear that the level of Sr^{90} contamination of milk will depend primarily upon the amounts of Sr^{90} and calcium in the ration. There would seem to be little gain in simply adding mineral calcium to an otherwise adequate diet. First, any dietary modification would have to be carried out continuously over a reasonably long time period to permit adaptation of the animal. Secondly, it would be difficult to get a diary cow to eat more than 200–300 g of calcium per day and higher levels might be inadvisable from the standpoint of the health of the animal.[21] Thus, if animals were normally consuming about 80 g of calcium per day, a reduction factor of from 2 to 3 is the most that could be expected by increasing the calcium intake on a usual diet.

It may be instructive to consider the sources of dietary calcium. For maximum effect one should in principle reduce the plant calcium to a minimum and increase the mineral calcium to a maximum. For purposes of discussion it is necessary to consider two phases of environmental contamination: (a) present conditions under which Sr^{90} contamination of plant material results in major part from surface contamination; (b) future conditions, if there are no further weapons tests, under which Sr^{90} has reached steady state in the biosphere and reaches the plant after incorporation in the soil and uptake by the roots.

At present, the levels of Sr^{90} in plant material will be determined largely by such factors as meteorological conditions and the surface and adsorptive capacity of the vegetative cover. Thus, the stable calcium and strontium contents of different plants give no guide as to degree of contamination expected.

In the future there will be an approach to constant relationships between Sr^{90}, stable strontium and stable calcium although it must be remembered that such relationships will depend upon reactions of Sr^{90} in the soil and plowing practices. Since it appears that the stable strontium to calcium ratio of most plant material is not greatly variable, then the normal calcium content of various forages can be used as an indication of future Sr^{90} contributions to the ration. For example, a ration consisting of 8 lb alfalfa hay, 12 lb timothy hay, and 12 lb of grain would supply a dairy cow with about 80 g of calcium; mineral supplementation to 160 g daily would be expected to give a reduction factor of about 2. On the other hand, a ration of 20 lb timothy hay and 12 lb grain would supply about 40 g of calcium per day and supplementation to 160 g would yield a reduction factor of about 4.

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A LARGE WELL CRYSTAL FOR THE DIRECT MEASUREMENT OF TRACE AMOUNTS OF RADIOISOTOPES IN ENVIRONMENTAL SAMPLES

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(*Received* 24 *June* 1960)

Abstract—A large diameter NaI(Tl) well crystal has been evaluated for use in the measurement of trace amounts of radioisotopes in environmental, farm produce, and bioassay samples. A 500-g sample in its plastic container is counted directly in the crystal. With this detector, about an order of magnitude greater sensitivity is obtained for most radioisotopes than was observed with a conventional 3-in. diameter by 3-in. thick crystal. The detection limits for the measurement of 15 radioisotopes which have been studied are about from 10^{-3} to 10^{-5} of the MPC's for these radioisotopes in drinking water. Typical spectra of vegetation, Columbia River water, and bioassay samples are presented.

INTRODUCTION

In the operation of the Hanford nuclear reactors and the associated chemical processing plants, small amounts of radioisotopes are released to the environs in the gaseous and liquid effluents. To determine the ultimate disposition of these radioisotopes, an extensive monitoring program, which includes the analysis of air filters, vegetation, farm produce, river water and well water from the plant environs, is carried out. The quantities of radioactivity present in these samples are very small, often less than the natural radioactivity, but it is essential to identify the isotopes in order to differentiate between plant emission, natural radioisotope, fallout or other possible sources, and in order to allow proper exposure estimation. Analyses of these samples were initially performed by wet radiochemical methods, but these methods were largely replaced 3 to 4 years ago by more sensitive γ-ray spectrometric techniques[1–3] which did not require chemical separations. For this work a 3-in. diameter by 3-in. thick NaI(Tl) detector multichannel analyzer system was used. With the availability of large NaI(Tl) crystals and techniques for obtaining good resolution[4] with them, it was apparent that a large improvement in sensitivity could be obtained.

To study the applicability of a large well crystal for the measurement of radioisotopes in environmental samples, a crystal was obtained which would hold a 500-ml polyethylene sample bottle. The dimensions of the crystal were $9\frac{3}{8}$ in. in diameter by $8\frac{1}{2}$ in. high, with a 3-in. diameter well extending to 3 in. from the bottom of the crystal. The crystal was beveled at 45° from a height of 6 in. to the top.

RESOLUTION

Resolution, which is defined as the width of a photopeak at one half its maximum counting rate, is a measure of the ability of a detection system to differentiate between γ-rays of different energies. Good resolution is essential in the γ-ray spectrometric analysis of complex mixtures of radioisotopes such as the radioisotopes on vegetation which result from fallout. Therefore a photomultiplier–crystal arrangement which would provide the best resolution was desired. Work at the Argonne National Laboratory[4] has shown that a cluster of 3-in. tubes viewing a large crystal provided far better resolution than that obtained by viewing the crystal with the single large photomultiplier tubes that were available.

* General Electric Company, Richland, Washington. Work performed under Contract No. AT(45-1)-1350 for the U.S. Atomic Energy Commission.

Viewing our large crystal with a cluster of seven 3-in. DuMont 6363 photomultipliers (with their individual outputs balanced by high voltage adjustment and their signals summed) provided a resolution of about 11.5 per cent for the width at half-maximum of the Cs^{137} (0.66 MeV) photopeak. Resolution measurements with 16-in. DuMont K-1328 and 12-in. EMI 9545 phototubes provided resolutions of about 16.5 per cent and 15 per cent, respectively. The use of a 2-in. thick lucite light pipe between the crystal and these tubes did not provide a significant improvement. Photographs of the final crystal–photomultiplier arrangement are shown in Fig. 1. The stainless steel surface separating the photomultiplier faces is coated with white paint (Plasite, Number 7100-B, Wisconsin Protective Coating Company, Green Bay, Wisconsin) for light reflection. The assembled crystal–photomultiplier system was housed in a 4-ft by 4-ft by 4-ft lead shield with 4-in. thick walls, and the output pulses from the detector were analyzed with a 256-channel energy analyzer.

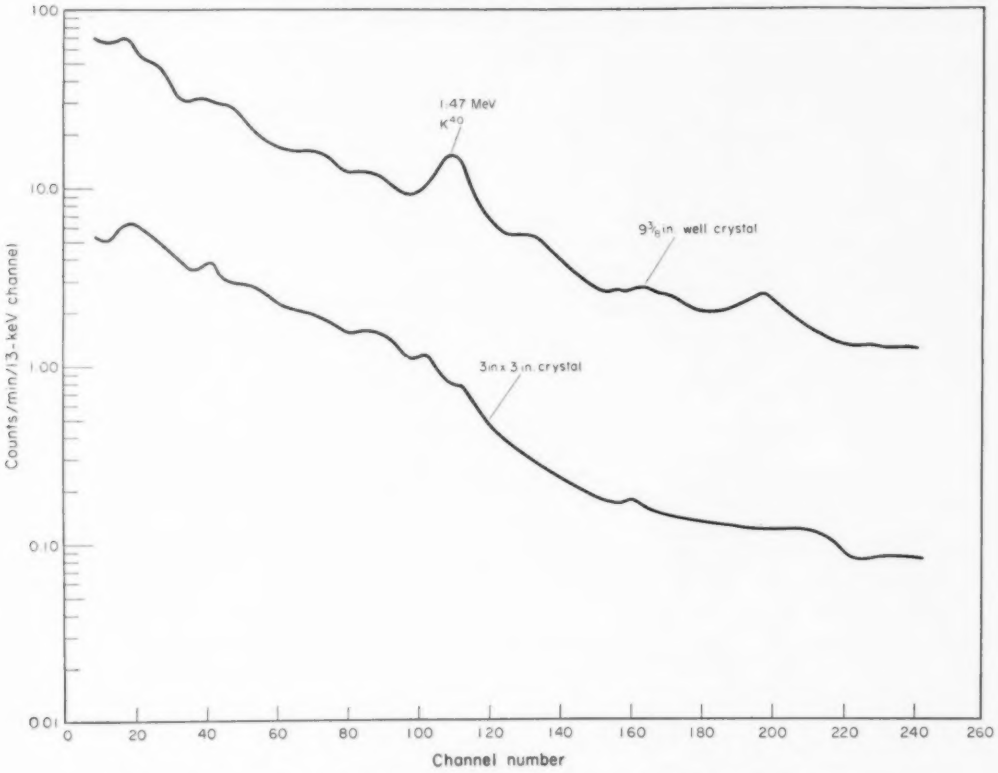

FIG. 2. Comparison of the background counting rates of the 3 in. and $9\frac{3}{8}$ in. crystals.

BACKGROUND MEASUREMENTS

The background counting rate of the large well crystal is compared with the background of a 3-in. by 3-in. crystal in Fig. 2. The large crystal background is, of course, much higher because of its larger volume. Its background could be reduced substantially by replacing its glass window with low-potassium glass or quartz and removing the sockets from the photomultiplier tubes. It is planned to make these changes as time permits. In spite of this high background, a much better sensitivity is available

FIG. 1(a). Crystal and photomultiplier units (face view).

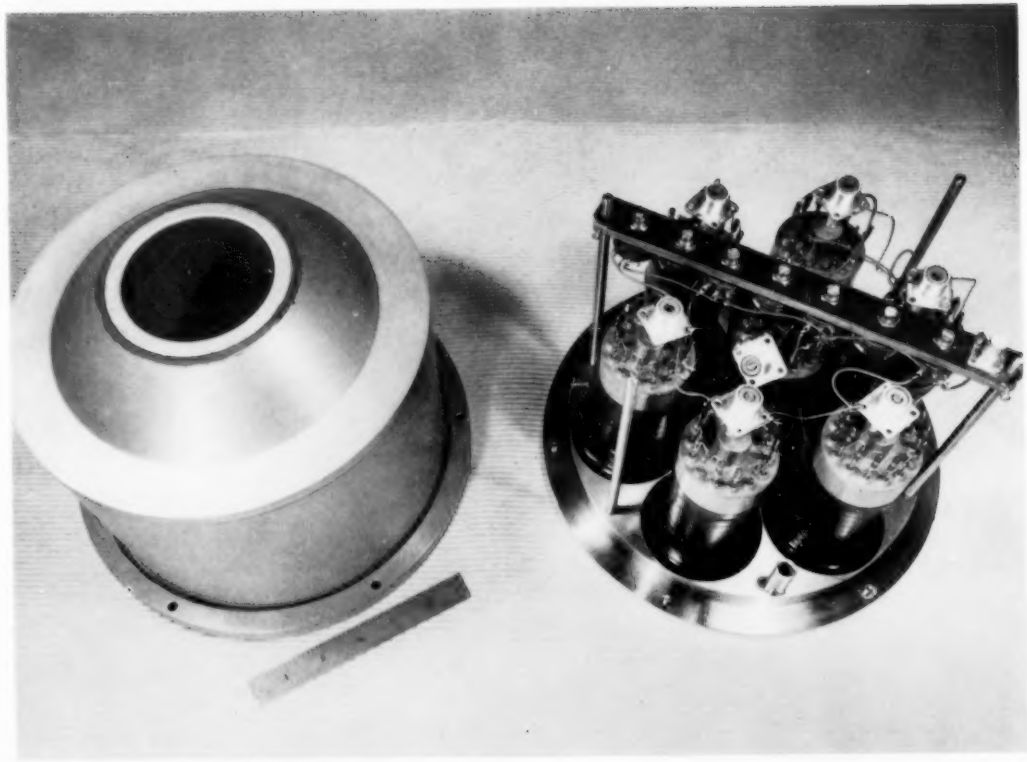

FIG. 1(b). Crystal and photomultipler units (end view).

with the large crystal than with the 3-in. by 3-in. crystal.

RADIOISOTOPE DETECTION LIMITS

The crystal was calibrated by counting water (or slightly acid) solutions of standard "spikes" in the 500-ml polyethylene sample bottles. When the radioisotopic concentration of a sample is measured from its γ-ray spectrum, the detection limit for each radioisotope depends on the concentrations of the other radioisotopes present, as well as on the detector counting efficiency, its background counting rate, the length of the count, etc. Thus, it is not possible to calculate a detection limit for a radioisotope which is applicable to all types of samples. For this reason, only detection limits for specific types of samples are reported here. Four detection limits (95 per cent confidence level) have been calculated for fifteen radioisotopes of interest. These "limits ' are all based on 50-min sample counts, a 50 or 1000 min background count and the condition that a single radioisotope, or one isotope plus the average amount of K^{40} in typical farm produce, is present. An area which represents approximately 80 per cent of the central portion of the principal photopeak is used in the measurement of the radioisotopes and for these calculations. These detection limits and the conditions used to calculate each of them are given in Table 1, along with the energies of the principal photopeaks. Also included in this table are the MPC values of the radioisotopes[5] in drinking water or foods.

It is apparent from these data that the sensitivities for the measurement of these isotopes are mostly from 3 to 5 orders of magnitude better than required to determine them at their MPC's, and would thus be entirely adequate for most monitoring surveys.

To illustrate the improvement in sensitivity with this large detector system over that available with a 3-in. by 3-in. crystal, the sensitivities of these two systems for the measurement of Zn^{65} and Cs^{137} are compared in Table 2.

Table 1. *Detection limits for* 500-g *samples* ($\mu\mu c/g$)
(95 per cent confidence level)

Isotope	Photopeak energy	I	II	III	IV	MPC
Na^{24}	2.75	0.018	0.013	0.018	0.013	2000
Sc^{46}	2.00*	0.019	0.014	0.019	0.014	400
Cr^{51}	0.325	0.11	0.083	0.12	0.085	20,000
Mn^{54}	0.845	0.018	0.013	0.018	0.013	1000
Co^{60}	2.51*	0.021	0.015	0.021	0.015	500
Cu^{64}	1.02*	0.10	0.073	0.10	0.076	200
Zn^{65}	1.12	0.027	0.019	0.027	0.020	1000
As^{76}	0.561	0.040	0.029	0.040	0.030	200
$ZrNb^{95}$	0.722 to 0.765	0.016	0.012	0.016	0.012	600 and 1000
Ru^{106}	0.515	0.094	0.068	0.095	0.069	100
I^{131}	0.364	0.017	0.012	0.017	0.012	20
Cs^{137}	0.662	0.014	0.010	0.014	0.010	200
$CePr^{144}$	0.145	0.13	0.097	0.14	0.10	100
Np^{239}	0.106	0.013	0.0096	0.014	0.010	1000
K^{40}	1.46	0.18	0.13	—	—	

I. 50-min count, 50-min background, only one radioisotope present.
II. 50-min count, 1000-min background, only one radioisotope present.
III. 50-min count, 50-min background, 3.6 $\mu\mu c/g$ (0.43 per cent K) K^{40} present.
IV. 50-min count, 1000-min background, 3.6 $\mu\mu c/g$ (0.43 per cent K) K^{40} present.

* Energy of the sum peak.

Table 2. Comparison of sensitivities available with the $9\frac{3}{8}$-in. well crystal and the standard 3-in. crystal with 500-g samples

($\mu\mu$c/g of sample, 95 per cent confidence level)

Radioisotope	Large crystal	Standard crystal*
Zn^{65}	0.027	0.36
Cs^{137}	0.014	0.16

* 500 ml polyethylene sample bottle on top of crystal.

It is apparent that the large crystal provides an improvement in sensitivity of about a factor of 12 for Zn^{65} and Cs^{137}. The improvement in sensitivity for other radioisotopes is about the same.

Another comparison of these two detectors is their relative counting efficiencies for the 500-g samples. The ratios of the observed counting rate with the large crystal to that with the 3-in. by 3-in. crystal for Cs^{137} and Zn^{65} are 35.8 and 40.1, respectively. If the large crystal had a background counting rate equal to that of the small crystal, the improvement in sensitivity of the large crystal–sample system over the standard system would approach these values.

The detection limits III and IV (Table I) are more realistic for vegetation or produce samples since they include any uncertainty caused by the presence of natural K^{40}. These detection limits will, of course, be influenced by the presence of other radioisotopes in the sample if they have higher energy γ-ray components and thus contribute "Compton background".

This substantial increase in sensitivity has greatly simplified and increased the precision of the measurement of radioisotopes in environmental and other "low-level" samples.

APPLICATIONS

Numerous applications of the large detector spectrometer have been made, and many others are apparent. At the present time, it is being used for the measurement of radioisotopes from reactor effluent water in the Columbia River, in aquatic life of the river and coastal waters, in farm produce from Columbia River irrigation projects, for radioisotopes in bioassay samples and for the measurement of fallout and/or plant released radioisotopes on vegetation. Sample preparation simply involves filling a 500-ml polyethylene bottle for liquid, or packing the bottle (with the top cut off for ease in loading) with a hand press to a density of about one for vegetation samples.

The spectrum of a typical 500-g vegetation sample from the environs of Hanford which was measured during November, 1959 is shown as Fig. 3. Even with this large crystal, a relatively low counting rate is obtained. With a counting rate of from 2 to 3 per cent of this, obtainable with a 3-in. by 3-in. crystal, a precise radioisotopic analysis would be very difficult.

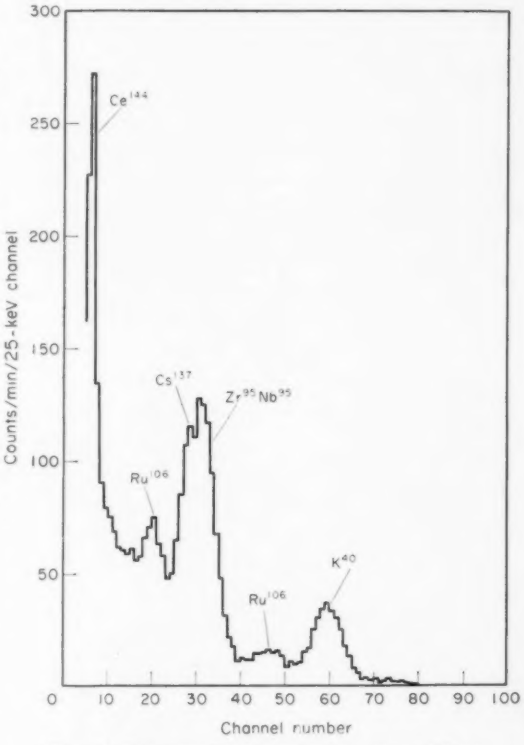

FIG. 3. Gamma-ray spectrum of a 500-g vegetation sample measured in the $9\frac{3}{8}$ in. well crystal.

Measurements of the γ-ray spectrum of a 500-ml sample of Columbia River water are shown in Fig. 4 and 5. This sample was collected about 30 miles below the Hanford reactors and these spectra were measured the day of sample collection, and after a 1-week decay period. The concentrations of Na^{24}, Cu^{64} and As^{76} are measured from the initial spectrum, while the concentrations of Zn^{65}, Cr^{51} and Np^{239} are measured from the second spectrum. The method of calculating these radioisotope concentrations from the

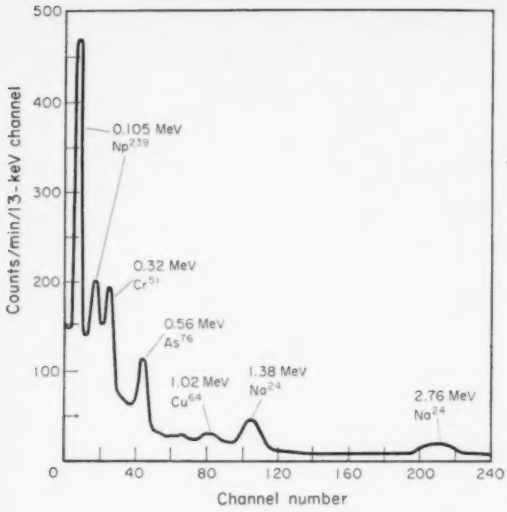

FIG. 4. Gamma-ray spectrum of a fresh Columbia River water sample.

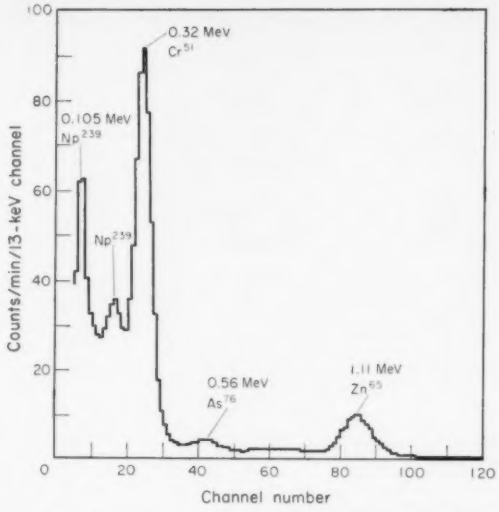

FIG. 5. Gamma-ray spectrum of a week-old Columbia River water sample.

γ-ray spectra was described earlier.[3] In view of the relatively low counting rate obtained, it is again apparent that a precise measurement of "low level" samples such as these would be difficult with smaller crystals.

Fig. 6 shows the γ-ray spectrum of a typical 24-hr urine sample (actually about 1500 ml evaporated down to 500 ml). It is apparent that the detector system is sensitive enough to allow a measurement of the natural potassium and the "fallout" Cs^{137} in the sample. The actual sensitivities for radioisotopes in urine samples are about one-third those in Column III of Table 1 (for 1500 ml samples evaporated to 500 ml). It is interesting that these values are, in general, from one to two orders of magnitude more sensitive than those recently reported for a radiochemical separations procedure for radioisotopes in urine.[6]

DISCUSSION

The large NaI(Tl) well crystal has proven to be an extremely useful tool in the analysis of "low level" environmental and bioassay samples. In many cases, it permits the direct measurement of radioisotopes which would otherwise require long and tedious chemical separations or concentration steps. The sensitivity for most radioisotopes (using the standard 500-g samples) is about a factor of 10 better than that available with a 3-in. by 3-in. crystal.

By counting a sample inside a large crystal, one is approaching the ultimate in counting efficiency and sensitivity. There is, however, considerable room for improvement in sensitivity. If the background could be reduced by a factor of 10, this would provide an approximate

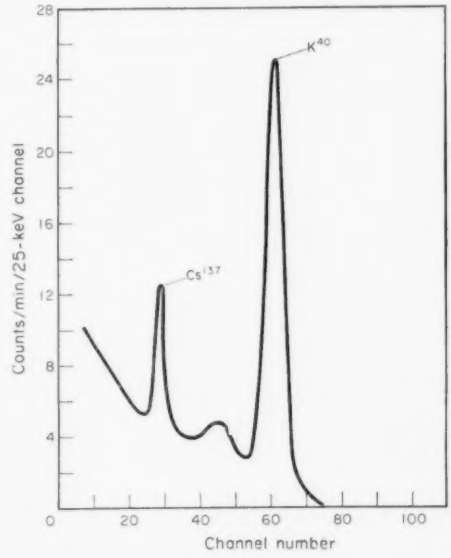

FIG. 6. Gamma-ray spectrum of a 24-hr urine sample.

threefold improvement in sensitivity. The possibility of reducing the background of a large crystal such as this, by use of a combination of low-background housing materials and anticoincidence shielding, is being studied.

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LOW-LEVEL POLONIUM AND RADIOLEAD ANALYSIS*

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Abstract—Methods are described for low-level Po^{210} and Pb^{210} analysis of amounts up to 1 l. volume of urine, 10–20 g of bone, 50–60 g tissue. The methods are applied to the analysis of experimental animals exposed to natural levels of radon and its short-lived daughters, yielding a natural Po whole body content of about 5×10^{-15} c/g. When the experimental animals are exposed to elevated daughter-free radon atmospheres, the Po and Pb body contents are increased. The study is part of a program to explore the usefulness of measurement of the Po content of miners' urine as an indicator of the integrated dose from radon and its daughters.

INTRODUCTION

In recent years, increased mining and processing of uranium has subjected numerous individuals to potentially hazardous concentrations of radon and its short-lived daughters.[1–3] There exists a widespread, but not unchallenged, contention, based on data[4] collected in the Joachimsthal and Schneeberg mines, that lung cancer may be caused by prolonged exposure to high levels of these radioactive substances. This paper describes a method for estimation of individual exposure which, correlated with measurement of air levels of radon and its daughters, may be used to relate with the future incidence of late lung disease and thereby permit a more exact evaluation of the hazard to man.

The radon hazard as presently understood[5–6] depends on the daughter product concentration in the air breathed. A large fraction of the aerosol-borne daughters deposit in the lung and there decay to the 19.6-year half-life Pb^{210}. Since the lead isotope deposits in the skeleton, has a long radioactive half-life, and is only slowly excreted, it might be expected that this lead reservoir would be a measure of the integrated radon exposure. Because Pb^{210} decays to the 5-day half-life Bi^{210}, which in turn decays to the 138-day half-life Po^{210}, the analysis for polonium in the urine of man could be[7] a measure of the lead stored and thus of the radon exposure. This paper describes a method for the analysis of small amounts of Po in large volumes of urine and tissue.

ANALYTICAL METHODS

After survey of the methods cited in the literature,[8–14] the following procedures were developed. For a more detailed description see Ref. 15.

Urine

Collect urine in a glass container. Add 25 ml concentrated HCl per estimated liter, or less, of urine and measure the volume. Transfer to a 1-l. Erlenmeyer flask for digestion and add 10 per cent by volume of concentrated HNO_3. Boil down to 40–50 ml and add 50 ml of 70 per cent $HClO_4$. Cover with a Speedy-Vap glass and continue boiling until white fumes appear. Add a few drops of HCl or HCHO to the solution occasionally and when no nitric oxide fumes appear, indicating that the excess nitric acid has been vaporized, remove cover and boil down to minimal volume. Cool and add 100 ml water, then neutralize with NaOH. Add HCl to make the solution 0.3–0.5 N, cover with watch glass and reflux for 2 hr. Transfer while hot to a plating column over a clean Ag foil (1.25 in. diameter circular area exposed), using three rinses of 0.5 N HCl, and add 200 mg ascorbic acid. Dilute to 200 ml with 0.5 N HCl. Place in a water bath (90–100°C) and using glass stirring rods, formed to give a slight propeller action, stir at 300–600 rev/min for 2.5 hr.

* This report is based on work performed under contract with the U.S. Atomic Energy Commission at the University of Rochester, Atomic Energy Project, Rochester, New York.

† Now at St. Mary's Hospital, Grand Junction, Colorado.

Our recovery with this procedure was 93.2 ± 4.8 per cent, for polonium added to 500–700 ml urine samples, sixteen samples. A technician had an average recovery of 87.3 ± 3.8 per cent for twenty-three similar samples.

Tissues

Add 2 ml concentrated HNO_3 per g tissue up to 25 g and 1 ml/g for larger samples. All weights should be wet weight and not more than 20 g of bone should be used in one sample. Let stand for several hours, preferably overnight. Heat gently and control foaming. When all solids have dissolved, cool and if much fat is present, extract with petroleum ether. Add 30–50 ml ether, swirl, remove supernate carefully with pipette and repeat the procedure twice. Extraction speeds digestion time. Boil down to small volume, add 25–30 ml of 70 per cent $HClO_4$ and continue treatment as for urines above.

Our average recovery from seventeen samples including various amounts and types of soft tissue and bone, to which polonium had been added, was 96.8 ± 2%.

Radiolead

To measure the amount of Pb^{210} in the sample, store for several weeks the solution from which Po has been plated, heat, transfer to plating column, add 100 mg ascorbic acid, and replate. The Po obtained on a second plating will be a measure of the amount of Pb contained in the solution. If small amounts, at or near naturally occurring levels, of Pb are anticipated up to 3 months between platings may be necessary to permit measurable amounts of polonium to grow into the solution.

COMMENTS ON METHODS

In working out this method, the following factors were determined.

(1) Decreased recoveries occurred when:
(a) foils were more than slightly discolored after plating;
(b) plating solution contained undissolved solids;
(c) HNO_3 was present in plating solution;
(d) ferric ion was present; ascorbic acid reduces it to ferrous ion which does not lower recovery;
(e) copper foil was used instead of Ag; Ni foil gave almost as good recovery as Ag but became discolored more easily.

(2) Recovery was most consistent with 2.5-hr plating time and when sides of column were rinsed down every hour with 0.5 N HCl.

(3) Recovery was as good with 1 $\mu\mu$c activity as with higher activities (Table 1).

(4) Five aliquots from the pooled urine of polonium injected rats were acidified and plated without prior digestion. The results showed an average recovery equal to 83 per cent of the recovery obtained when an equal five aliquots from the pool were treated as described above.

Recovery percentage was determined, in those cases where added Po was used, by depositing known amounts of a stock Po solution on Ag foils and allowing them to air dry.

Table 1. *Recovery of* Po *from* 0.5 N HCl

Number of samples	Added activity ($\mu\mu$c)	Recovered activity ($\mu\mu$c)	% recovered
1	79.3	77.8	98.2
1	32.3	29.8	92.6
1	15.6	14.6	93.7
1	7.28	6.95	95.5
1	3.27	3.57	105
1	3.12	2.65	85.1
2	2.02	1.84	91.2
1	1.04	0.97	92.4
2	0.675	0.656	97.3

The α-activity of the foils was measured by counting with an α-scintillation counter. The efficiency of the counter was 45 per cent for Po α's as determined by counting three different Ra D, E + F sources which had been calibrated by National Bureau of Standards. The background counting rate of the instrument was 4–6 counts/hr.

The method described above is essentially that developed by FINK *et al.* at the University of Rochester Atomic Energy Project but had to be tested and modified for recovery of $\mu\mu$c amounts of Po in various samples. Other methods which were either more simple or faster proved unsatisfactory for the stated purpose.

Once the method had been worked out satisfactorily, it was applied to analysis of various tissues and to whole animals. Analysis of mice and rats for naturally occurring amounts

of Po was attempted first. Later, samples of human tissues were obtained at autopsy from persons presumably exposed to only natural atmospheric levels of radon. Finally, samples from animals exposed to radon in a special chamber as well as two bats* which had been trapped in a uranium mine with a high radon content were analyzed.

RESULTS OF TISSUE ANALYSIS

Natural levels of polonium

Since a body burden of about 10^{-10} g of Ra has been found in man and since the atmosphere contains of the order of 10^{-13} c/l. of radon, plus its daughter products, it might be expected that small amounts of Pb would be found in all humans and animal species even though there may be no history of special exposure. This natural Pb level would then form the background upon which mine radon exposure connected Pb would be superimposed. Table 2 lists the results of Po analysis of unexposed tissues of various sorts. It is seen that a small amount of Po can indeed be found, and there is a suggestion that the concentration of Po in bone is somewhat higher than in soft tissues. In each set of experiments represented in the table, polonium-spiked samples were used to verify recovery (91–96 per cent) and reagent blanks were analyzed. Analyses of the tissues were in all cases performed shortly after death and specified the Po^{210} content only.

Table 2. Natural polonium content

Species	Tissue	Weight (g)	Activity (dpm)	Activity ($\mu\mu$c/g)
Mouse 1	whole body	25	0.38	0.0068
Mouse 2	whole body	25	0.36	0.0065
Mouse 3	whole body	25	0.34	0.0061
Mouse 4	whole body	25	0.30	0.0054
Mouse 5	whole body	25	0.21	0.0038
Mouse 6	whole body	25	0.27	0.0049
Rat 1	whole body	150.4	1.69	0.0051
Rat 2	whole body	137.7	1.42	0.0046
Man A_1	rib and sternum	42.4	3.04	0.032
Man A_2	rib and sternum	44.5	3.02	0.031
Man B_1	rib and sternum	46.6	7.80	0.075
Man B_2	rib and sternum	33.8	0.90	0.012
Man B_3	rib and sternum	37.5	1.18	0.014
Man C_1	ribs	9.3	0.0	0.00
Man C_2	ribs	11.5	0.093	0.0036
Man D_1	lung	49.5	0.345	0.0031
Man D_2	lung	52.0	0.690	0.0060
Man D_3	lung	53.6	0.389	0.0033

* Species *Corynorhinus Rafinesquei*, supplied through the courtesy of Dr. MITCHELL R. ZAVON, U.S. Public Health Service.

Polonium in whole animals after exposure to high radon levels

Table 3 lists the results of Po analyses of whole animals exposed to radon as indicated. Except in the case of the bats caught in the uranium mine, the exposure was to filtered, rapidly circulated, radon-containing air so that the air as breathed was substantially free of radon daughter products. The daughter products were produced by the tissue-dissolved radon and, to a small extent, by the radon in the lung and its passages. In the set of two rats sacrificed immediately after exposure and analyzed 246 days later, the polonium detected is that formed after death with no opportunity for excretion. The activity detected was 82 per cent of that predicted by calculation.[15]

The polonium level in the exposed mice, due to the long time intervening between exposure and analysis, should be equal to the Pb^{210} content which in this case is diminished by the opportunity for excretion during life and by the initial blowing-off period which reduces the tissue radon content. As a consequence of these factors, the Po whole-body content is much lower for the mice than the rats. The activity detected was about 92 per cent of that which was calculated,[15] assuming no excretion during life.

The bats, caught in the uranium mine and presumably exposed to high natural levels of radon plus daughters, yield an expected high Po body content which, because of the long interval between sacrifice and analysis, approximates the body Pb content.

Bone levels of Po^{210} *and* Pb^{210}

Table 4 lists the result of an experiment exploring the bone deposition of Po and Pb. Nine mice were exposed to 200 μc/l. of daughter-free radon for 15 hr and permitted to live until

Table 3. Radon derived polonium in whole animals

Species	Weight (g)	Radon exposure*	Activity ($\mu\mu$c)	Activity ($\mu\mu$c/g)	Remarks
Rat	187	0.438	39.6†	21.2	Sacrificed immediately
Rat	202	0.438	40.3†	20.0	Analyzed 246 days later
Male mouse	15.7	1.0	7.5	0.48	Died natural death
Male mouse	19.6	1.0	9.38	0.48	Analyzed 791 days
Male mouse	19.1	1.0	9.22	0.48	after exposure
Male mouse	14.3	1.0	9.29	0.65	
Male mouse	16.6	1.0	11.93	0.72	
Female mouse	16.9	1.0	11.79	0.70	Died natural death
Female mouse	16.3	1.0	11.50	0.70	Analyzed 774 days
Female mouse	13.7	1.0	9.74	0.71	after exposure
Female mouse	8.7	1.0	10.42	1.20	
Female mouse	17.6	1.0	5.54	0.32	
Bat	(25)	—	14.7	(0.59)	1 yr between death and analysis; weight was
Bat	(25)	—	63.7	(2.55)	estimated

* mc hr/l. of daughter-free radon, e.g. 5-hr exposure to concentration of 200 μc/l. or 8 hr to 125 μc/l.
† An aliquot of 1 per cent of total digest was analyzed.

death which occurred from 90 to 100 days after exposure. A group of six mice were analyzed for Po whole body content, and the Po-deprived

Table 4. Tissue distribution of polonium and lead in mice 90–100 days after exposure for 15 hr to 200 μc per l. of radon

Sample	Weight (g)	Po^{210} activity ($\mu\mu$c)	Po^{210} activity ($\mu\mu$c/g)	Pb^{210} activity ($\mu\mu$c)	Pb^{210} activity ($\mu\mu$c/g)
Average whole body, 6 mice	17.4	62.7	3.61	124.0	7.12
Mouse A soft tissue	16.6	14.5	0.87	47.8	2.88
Bone	0.85	22.7	26.7	78.8	92.8
Mouse B soft tissue	27.1	36.9	1.36	35.2	1.30
Bone	0.51	9.7	19.1	62.8	123.0
Mouse C soft tissue	19.1	34.2	1.78	49.7	2.59
Bone	0.83	12.4	14.9	114.0	137.0

plating solution was stored for a sufficiently long time to enable a measurement of the Pb content from newly formed Po.

Three additional mice were separately treated by heating the carcass in 3 per cent KOH–ethylene glycol solution to dissolve the soft tissues, permitting the filtering-off of the bones and a separate analysis of the two fractions for Po and Pb.

In all cases the interval between death and the first plating of Po was about 30 days. The results entered in the table have been corrected so that they correspond to the tissue content at the time of death. It is found that the Po whole body content is about 40 per cent of the Pb whole body content. The concentration of Po and Pb in the bone is many times higher than in soft tissue. For intravenously injected Po, the per cent body content per g bone is about half that for average tissue. The present investigation creates the strong presumption that tissue distribution and probably excretion data for Po as established by Po injection studies may not be used with confidence in the calculation of the fate of Po as a daughter product of lead.

If the amount of whole-body Po in this animal group is compared with the mouse data in Table 3 and adjusted for exposure, the reduced Po

levels of the latter group, caused by the longer interval between exposure and death, permit the estimate that the Pb body content had decreased by about half in the 770 day post-exposure period.

SUMMARY

Methods were developed and tested for the quantitative analysis of liter volumes of urine, 10–20 g bone, and 50–60 g soft tissue for polonium content in the $\mu\mu$c range. Radiolead content can be determined by saving the solutions and reanalyzing for the Po which has formed from the radiolead.

By using test animals exposed to radon, it has been shown that radiolead accumulates in the animals, particularly in the skeleton, and that it has a relatively long effective half-life, in mice.

Data corrected to time of death indicate the Po content of the animals investigated is about 40 per cent of the Pb content, and since both decrease with time after exposure, a measurement of excreted Po may be an indication of stored radiolead, which in turn is related to the radon exposure of the animals.

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AN ANALYSIS OF THE CHICAGO SYNCHROCYCLOTRON PERSONNEL DOSES OVER A 6-YEAR PERIOD*

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Abstract—This report is an analysis over a 6-year period (1954–1959). Included are: history and operational data of the Chicago Synchrocyclotron, comparative dose data by groups and years, and radiation incidents. In addition, doses greater than 0.3 rem/week are correlated with their causes and major operations are ordered according to their dose-producing potential.

The aim is to provide a basis for dose estimates and work schedule construction, as well as to present a view of the Synchrocyclotron from a Health Physics standpoint.

In general, the average yearly dose per man has tended to decrease, within the frame of current maximum permissible limits. Weekly overdoses have been greatly reduced in 1958 and 1959.

INTRODUCTION

THIS report,† an analysis of personnel doses at the University of Chicago Synchrocyclotron over a 6-year period,‡ has a twofold aim. First, it is an attempt to present a clear picture of the machine itself and the doses incurred by its personnel; its second and primary aim is to correlate personnel doses with their causes in the maintenance and operation of the machine. These two complementary goals are realized in Parts I and II, respectively, and are meant to serve as an aid to the health physicist, by providing a basis upon which to estimate the doses to be expected from a given operation.

The ability to estimate accurately personnel doses, coupled with a sound understanding of the workings of the machine, enables the health physicist to control the doses by means of well-planned work schedules. That is, he is able to recommend distribution of men and working time so that doses are kept low, while maintenance time is not unduly extended.

Such work schedules are indeed necessary in the case of a high-energy accelerator, a complex device operating on a 24-hr/day, 7 days/week basis. Here, maintenance is both frequent and

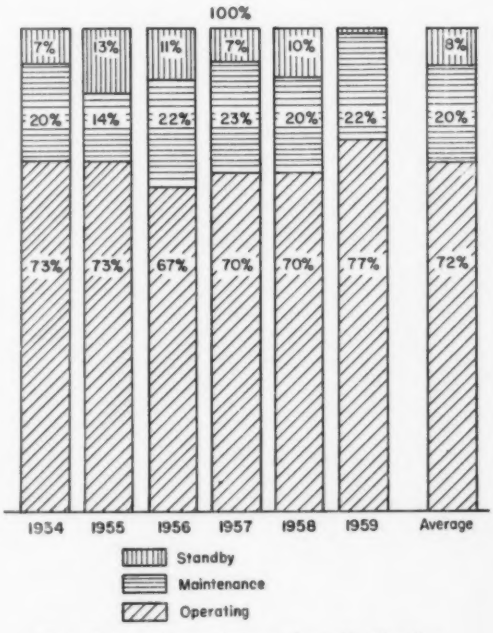

FIG. 1. Synchrocyclotron time distribution.

* Research supported by a joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

† A condensation of EFINS Report No. 348, under the same title.

‡ From January 1954 to December 1959.

diverse, and is usually performed in an atmosphere of urgency, under pressure from the experiment in progress.

BASIS

(1) The computations are based on the general *weekly* dose records filed at Argonne Cancer Research Hospital.

Due to certain difficulties involved in handling the ACRH early dose records (1951–1953), this statistical analysis only covers the period from the beginning of 1954 to the end of 1959 (or two-thirds of the lifetime of the Accelerator), which has been judged sufficiently long since no emphasis on absolute totals of personnel doses is necessary.

(2) Personnel working with, or for the Accelerator have been classified in four groups:

(a) "operator" (control and electrical maintenance);
(b) "crew" (mechanical maintenance);
(c) "research" (RG1-6; Health Physics-HP and Medical Physics-MP);
(d) "others" (Administration-A, Drafting Dept.-D, Machine Shop-M, and Housekeeping Serv.-J).

In the case of the "operator" and "crew" groups, all past and present personnel have been considered to improve the statistics in the early years. This has not been judged necessary for the "research" group (more numerous) or "others" (less important from our standpoint), in which, only present personnel have been accounted for.

For simplicity and comparison facility, personnel in the four groups have been taken as working only integer numbers of calendar years. This means that a certain number of doses received during partial calendar years (at the beginning and/or termination of individual's employment terms), have been neglected. However, the relative error: (1 — (dose used/total dose)) = 12 per cent, introduced on the average by this simplification, has been compensated (see "errors" below). So the term "present" applies to the individuals whose term of employment has included, at least, the entire year 1959 (from 1 January to 31 December).

(3) In general, doses-in-file are recorded as almost exclusively due to γ-radiation. Electron as well as nucleon contributions seldom occurred except in the case of the "operator" group, with a component (in rems) of about 30 per cent of fast neutrons and/or protons (but only monitored since 1957).

The dose-unit consistently used throughout this report is the rem, i.e. unit of absorbed dose (rad = 100 ergs, or 6.24×10^7 MeV, per g of irradiated material), corrected for the relative biological effectiveness in case of high-energy nucleons (RBE = ratio (γ-dose/nucleon-dose) to produce γ-equivalent biological effect; it has been conservatively taken as 10).

Doses are regarded as whole-body doses. (The very few recorded in file as hand-doses have not been computed). The term "exposure" is used as the period of time during which a person has been subjected to radiation, where as "number of exposures," is used as the number of times the person has absorbed a substantial dose (greater than 0.3 rem) while performing a set of operations in a radiation area.

(4) During the 6-year period the DuPont dosimeter film packet, type 558 (film types 508 and 1290) has been invariably used as β-, γ- and thermal-neutron-monitor. It has been processed by the HP group at ACRH, with weekly calibrations against 100 kV peak X-rays (1 mm Cu, 0.25 mm Al filters), a Victoreen-chamber-calibrated Co^{60} source and an uranium slab.

Since 1957, the Kodak Neutron Film, Type A, has been included. It is processed by Landauer Jr. Co. on the basis of 40-hr exposure to 20 PuBe-n/cm^2 sec (or 2.88×10^6, 5-MeV-n/cm^2 40 hr) as equivalent to 0.100 rem.

(5) Information regarding the Synchrocyclotron itself (history, time distribution, maintenance operations, etc.) is based on:

(a) The "Navy Large Cyclotron Status Report", prepared since 1949 on monthly or quarterly basis.
(b) The "170-in. Synchrocyclotron Machine Operating Record" (the Accelerator's daily log).
(c) The "170-in. Synchrocyclotron Progress Report" I-IV, by H. L. Anderson, J. Marshall (1947–1952).
(d) Personal interviews. At this point, we are especially indebted to C. Bordeaux,

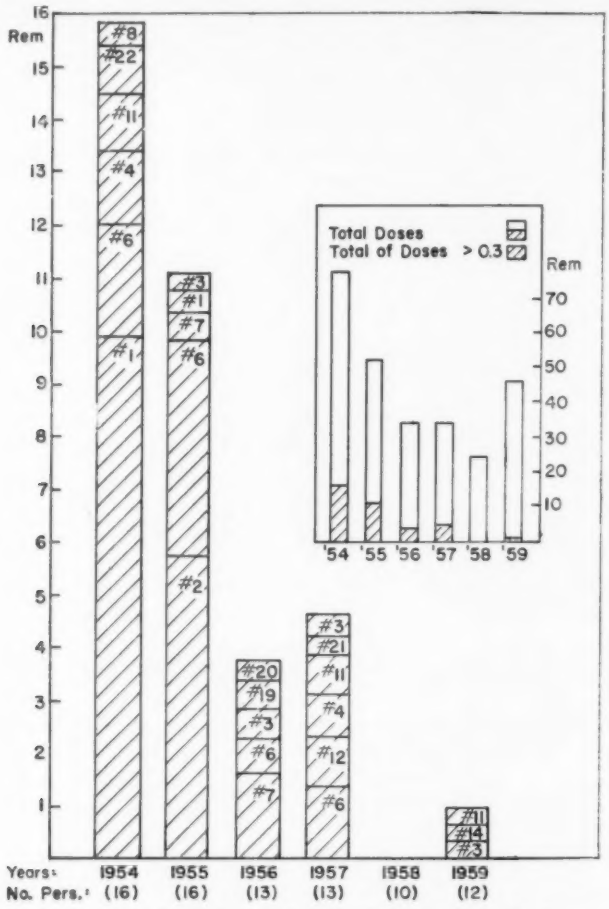

FIG. 2. Scale = 1.5 cm = 1.0 rem. Total of doses > 0.3 rem with causes by years. Group = "operator".

Chief Electrical Engineer of the Accelerator, for his co-operation and initiative. Without his valuable contributions this report would have been practically unrealizable.

(6) "Incidental exposures" have been excluded in the general computations. They are treated in a separate table, prepared with all the data available in our files.

ERRORS

Approximately 80 per cent of the number of recorded weekly doses lie in the lowest region (40 per cent for the operator group), below the minimum threshold sensitivity of the DuPont film monitors used, for 1.2 MeV. They are recorded in the general file as <25 mr. In order to obtain the best possible approach to the true doses in the considerations of this report, the following estimate was previously made to assign a standard value to these ambiguous readings:

Assuming that, on the average, the most probable dose for "25" is its mean value, or 12.5 mr, the gross average dose for the personnel and period considered amounted to 26.4 mr/man-week. Using this average, we estimated the

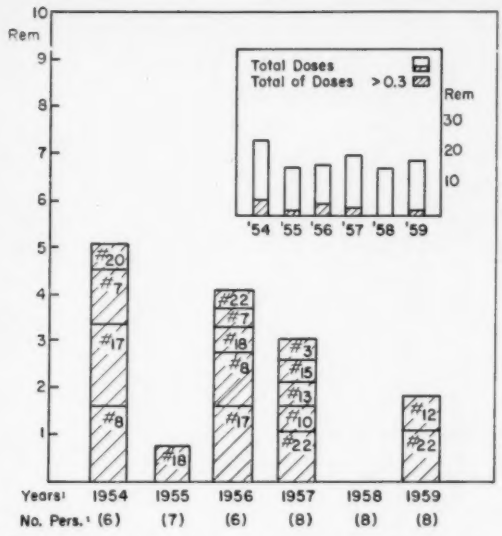

FIG. 3. Scale = 1.5 cm = 1.0 rem. Total of doses > 0.3 rem with causes by years. Group = "crew".

probable contribution due to non-recorded and non-considered doses as follows, (the total number of weekly doses considered is approximately 15,000):

(a) Film badges not worn, 10 per cent, or 2.1 mr/man-week.
(b) Films lost or destroyed, 2 per cent, or 0.5 mr/man-week.
(c) "Integer-years" simplification, 12 per cent, or 3.2 mr/man-week.
(d) Non-recorded doses due to other environmental sources*—about 6 mr/man week.

This gave a total of 23.8 mr/man week. Therefore, in order to compensate this value we chose to take $<25 = 25$ mr, throughout our computations.

Neutron doses contributed to only 30 per cent of the total "Operator" doses, 70 per cent of these exposures being recorded as <10 mrem. They only apply to less than the last half of the period considered. In this report, these relatively few "<10" doses are taken as 10 mrem. The same criterion (maximum value) has been followed for the "<50" mr γ-doses reported during about 10 months in 1954, somewhat compensating the lack of neutron data in this period.

The accuracy of the dose data in any case is limited by the photographic technique to about ± 10 per cent.

Acknowledgments—We wish to thank T. CRANE, P. DEPERRO and R. EHRLICH, of the Health Physics Group, for their very able assistance in collecting data, performing computations and for their many useful suggestions.

In addition to C. BORDEAUX, whose co-operation has been already mentioned, we are indebted, in general, to the staff of the Synchrocyclotron, and to the HP Group at Argonne Cancer Research Hospital.

PART I

Part I consists of two sections. The first presents some data concerning the operation of the Synchrocyclotron: an historical outline, time distribution and typical schedule.

The second section contains data on the doses received by Synchrocyclotron personnel.

Table 2 is an attempt to illuminate the general situation from several different angles, making possible easy comparisons at a glance.

It will be seen that the relative differences between the average dose/"high man" per week and average dose/man per week are similar for the groups, "operator," "crew", and "research", while the two quantities are very nearly equal for the "others" group. (Here, "high" week refers to the week of the year during which the total group dose was a maximum. The "high" man received the largest dose in his group during a particular year).

Tables 3 and 4 complement this data with summaries of accumulated dose data and radiation "incidents".

Synchrocyclotron data

Construction of the 170-in. University of Chicago Synchrocyclotron was begun in July, 1947, and the machine underwent its first experimental trial in February of 1951. Since then it has been almost continually modified and improved.

During the period from 1954 to 1959, which

* Neutron background, X-ray diagnosis, internal natural sources, fallout, etc., totaling about 25 rem/man 70 years (AEC data). Gamma-background (3.5 mr/week), already accounted in film badges on rack, was excluded.

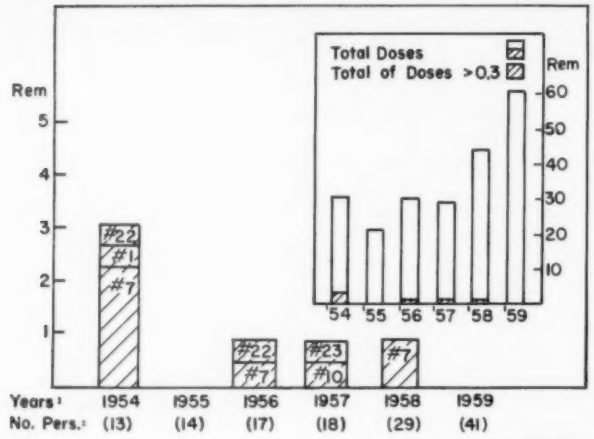

FIG. 4. Scale = 1.5 cm = 1.0 rem. Total of doses > 0.3 rem with causes by years. Group = "research".

FIG. 5. Scale = 1.5 cm = 1.0 rem. Total of doses > 0.3 rem with causes by years. Group = "others".

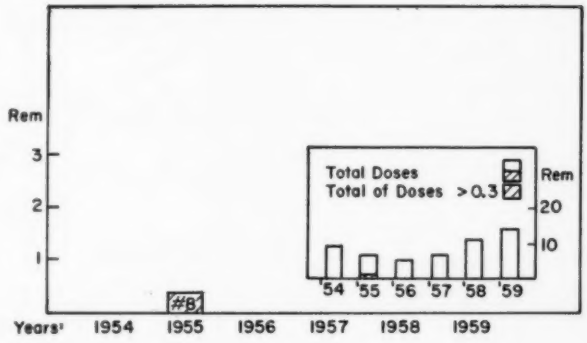

Table 1. *Typical synchrocyclotron schedule* (1959)

Group	Station			Period (weeks)
RG–2	Proton	10^{10} p/cm² sec	450 MeV	3
RG–7	Proton	10^{10} p/cm² sec	450 MeV	3
MP	Proton	10^{8} p/cm² sec	150 MeV	1
		Total consecutive weeks for proton experiments		= 7
RG–1	Meson	$(\pi - e)$		3
RG–5	Meson	(mm – parity)	60 MeV π^+ or π^-	
		10^2 π^+/cm² sec	or 50 MeV π^-	3
RG–6	Meson	$(\pi - \mu - e)$		
		10^4 π/cm² sec	60 MeV π^+ or π^-	3
		Total consecutive weeks for meson experiments		= 9

Cycle = 16 weeks, or three cycles during a year.

Each group is able to perform about 3 experiments per year.

Approximately 1 day (every 6 weeks), plus 1 week (every 25 weeks) is scheduled for maintenance work, or 6 per cent of the year. Therefore, about 14 per cent of the average year (or 70 per cent of the total main tenance) has been spent in unscheduled maintenance work.

Table 2. General dose data by groups and years

"Operator"	1954	1955	1956	1957	1958	1959
No. of persons	16	16	13	13	10	12
Total dose	77.42	52.04	34.03	33.68	23.49	45.20
Dose/man per "high week"	0.880	0.750	0.560	0.500	0.214	0.340
Avg. dose/"high man" per week	0.174	0.131	0.092	0.078	0.072	0.094
Avg. dose/man per week	0.093	0.063	0.050	0.050	0.045	0.072
"Crew"						
No. of persons	6	7	6	8	8	8
Total dose	23.17	15.31	15.87	20.23	14.83	17.24
Dose/man per "high week"	1.420	0.660	0.840	0.960	0.275	0.430
Avg. dose/"high man" per week	0.109	0.058	0.067	0.087	0.043	0.063
Avg. dose/man per week	0.074	0.042	0.051	0.049	0.036	0.041
"Research": RG1 to RG6, HP and MP						
No. of persons	13	14	17	18	29	41
Total dose	33.49	21.40	29.81	28.60	43.84	61.26
Dose/man per "high week"	0.560	0.900	1.500	0.480	0.880	0.240
Avg. dose/"high man" per week	0.087	0.044	0.067	0.065	0.049	0.043
Avg. dose/man per week	0.050	0.029	0.034	0.031	0.029	0.029
"Others": A, D, J and M						
No. of persons	4	4	4	5	8	10
Total dose	9.52	6.56	5.56	6.49	11.40	13.90
Dose/man per "high week"	0.560	0.330	0.140	0.124	0.250	0.220
Avg. dose/"high man" per week	0.052	0.042	0.028	0.027	0.035	0.031
Avg. dose/man per week	0.046	0.032	0.027	0.025	0.027	0.027

concerns us here, the following major developments were introduced (partial list):

1954: The "Hydra-target", a trolley car with twelve thermocouple–target assemblies, and radial–azimuthal remote controls, was put into use.

1955: The newly developed cold-cathode ion source was installed, replacing the old filament type. Protons were successfully deflected and polarized into the Meson Station.

1956: Protons were focused into the Meson Station. Construction of the Proton Station began.

1957: The Proton Station was completed and received the proton beam for experimental purposes.

1958: The beam into the Proton Station was successfully polarized and focused.

1959: The operational power limit of the Synchrocyclotron was increased to 32 W.

The power output from the Synchrocyclotron has been maintained at 20 W, except for a brief period in 1959, when 32 W were obtained.

PART II

*Correlations**

The results of the search made for the sources responsible for weekly doses greater than 0.3 rem are presented in this part.

It starts with the list of the twenty pertinent causes found.†

* Doses greater than 0.3 rem with causes.

† The numbers assigned to the causes read up to 23, but the numbers 5, 9 and 16 were rejected after the final correlations.

There follows a set of four figures which indicate for each group, the contribution of each cause to the total of doses >0.3 rem. In the insets of these figures, the total yearly dose, given in the data of Table 2, is compared with the total of doses >0.3 rem. Identical scales are used for facility of comparison.

of nearly constant data, and "average" to the estimated average of relatively variable data. The expression "beam-time" represents about 72 per cent of the actual time (see Fig. 1).

Fig. 7 provides an ordering of the major work from the standpoint of its dose-producing potential. Here, the doses per team are derived

Table 3. *Number of persons* (*P*) *in given ranges of yearly accumulated dose rate, by groups*
(*Y*) = number of years for which the ADR's apply.
All present personnel (by 31 January 1959)
Incidental doses included.

	Accumulated dose rate (ADR) in given range (rem)				
	1.0–2.0	>2.0–3.0	>3.0–4.0	>4.0–5.0	>5.0
"Operator"	2(*P*)–1(*Y*)	1(*P*)–3(*Y*) 1(*P*)–6(*Y*)	1(*P*)–1(*Y*) 1(*P*)–2(*Y*) 1(*P*)–3(*Y*) 1(*P*)–5(*Y*) 2(*P*)–6(*Y*)	2(*P*)–6(*Y*)	
"Crew"		1(*P*)–3(*Y*) 6(*P*)–6(*Y*)			
"Research"	10(*P*)–1(*Y*) 3(*P*)–2(*Y*) 1(*P*)–3(*Y*) 4(*P*)–4(*Y*) 1(*P*)–5(*Y*) 9(*P*)–6(*Y*)	2(*P*)–1(*Y*) 1(*P*)–2(*Y*) 3(*P*)–4(*Y*) 2(*P*)–6(*Y*)			
"Other"	1(*P*)–1(*Y*) 3(*P*)–2(*Y*) 2(*P*)–3(*Y*) 1(*P*)–4(*Y*) 3(*P*)–6(*Y*)				

A relative evaluation of each cause can be obtained by examination of Fig. 6. In it, the three intervening parameters: (1) total dose, (2) no. of persons, and (3) no. of exposures, are represented together, corresponding to each cause. An inset points out the years in which such causes are responsible for doses greater than 0.3 rem.

Finally, each cause is individually analyzed in "Data and Comments on Causes". In this analysis, the word "about" refers to the value

from the data in the analysis, by weighting each operation with respect to frequency, duration, dose rate in working area, and number of persons at work, under typical conditions.

Note: Although the data and correlations presented in this part are based on a scrupulous study of the available sources of information, the difficulties involved have been considerable, for no emphasis was placed on this area in the past. Therefore, this material should be regarded as an approach to the facts.

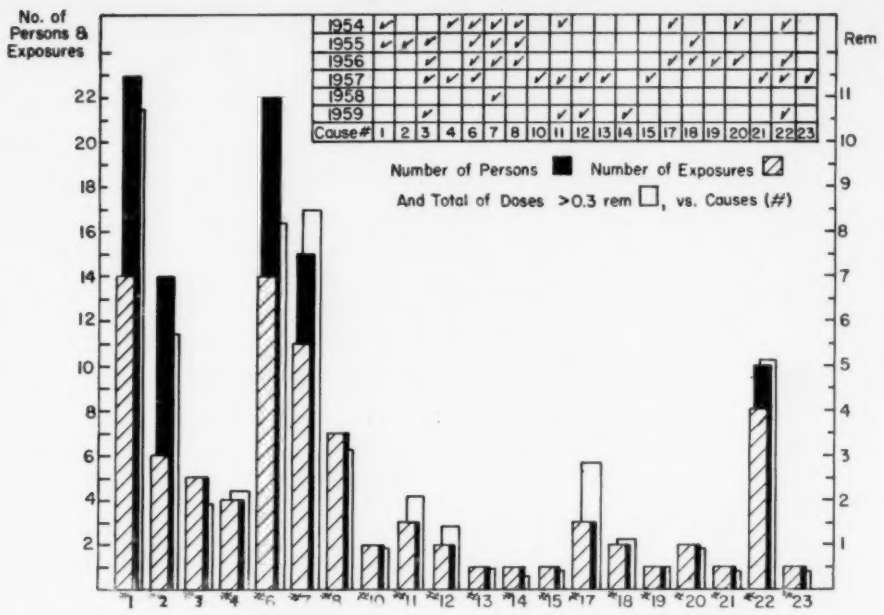
No. of Persons & Exposures
Rem
1954
1955
1956
1957
1958
1959
Cause# 1 2 3 4 6 7 8 10 11 12 13 14 15 16 17 18 19 20 21 22 23
Number of Persons
Number of Exposures
And Total of Doses >0.3 rem, vs. Causes (#)

FIG. 6. Number of persons and exposures, and total of doses >0.3 rem, vs. causes.

Causes of doses greater than 0.3 rem

No.	Cause
1	Filament ion source
2	Cold cathode ion source development
3	Cold cathode ion source
4	Vacuum leaks
6	Target maintenance
7	Extractors
8	Bearing replacements
10	Dee cleaning
11	Stub cleaning
12	Clearing system
13	Water cooling system
14	Oil condenser fires
15	Diffusion pump maintenance
17	Shielding
18	Plastic windows
19	Lights in tank
20	2-in. probe maintenance
21	Clipper changes
22	Indeterminate
23	Radiochemistry

DATA AND COMMENTS ON CAUSES

No. 1 *filament ion source*

Two interchangeable units were used between February 1951 and March 1955. Unit maintenance required frequent reversals and replacements, as well as occasional repair of water leaks, anode maintenance, and unit alignments. During 1954–1955, filaments were replaced after about 6 hr of beam time. This required one operator for approximately 15 min. Reversals were necessary at about 2 hr intervals, the procedure requiring one operator for about 2 min. The radiation level in the working area was about 0.1 r/hr.

No. 2 *cold cathode ion source development*

The present cold cathode ion source was developed during a 4-month period in 1954–1955. One or two operators worked on the ion source for about 2 hr/week, in a radiation level of 0.1 r/hr.

No. 3 *cold cathode ion source*

The cold cathode ion source has been in use since 1955. Major maintenance tasks include unit alignments (5 min time, once a week), electrode replacements (about 1 hr every 3 weeks), and improvements and/or repairs (average 1.5 hr every 2 weeks). These operations all require an average of two operators to work in a radiation level of about 0.2 r/hr.

Table 4. Incidental exposures (1951–1959)

DATE	0100 hours 8/25/1951.
PERS	Non-staff experimenter.
NOTF	Not stated (H.P. Report: 8/30/51).
CIRC	Experimenter entered the exp. area while beam on (high background but no safety interlocks installed) without requesting beam off from control rm. (established procedure).
BEAM	250-MeV deuterons. Beam intensity on target 0.1 $\mu\mu$A.
REMK	Direct beam dose rate equivalent to 1800 r of X-rays. Range in tissue about 10 in.
TIME	About 30 sec in exp. area. Unknown in direct beam.
BODY	Probably total body.
MONTR	Pocket chamber: negligible reading.
TESTS	None.
DOSE	Max. of 0.5 r (based on total body exposed in direct beam during 30 sec).
RECOM	One week away from radiation sources.
DATE	1530 hours 2/11/1955.
PERS	Staff physicist and visitor.
NOTF	Not stated (H.P. Report: 2/12/55).
CIRC	Beam turned on without any aural warning (standard practice), while visitor tour in exp. area.
BEAM	External protons. Intensity at ion. chamber in cyc. pit about 10^{-6} A.
REMK	Previous measured dose rate at 20 ft from beam in exp. area = 0.375 rem/hr.
TIME	Max. 60 sec at 15–40 ft from path of beam.
BODY	Total body.
MONTR	None.
TESTS	None.
DOSE	Max. of 0.625 rem, fast neutrons, per individual.
RECOM	Restatement of standard safety procedures.
DATE	1525 hrs 9/13/1955.
PERS	Staff physicist and visitor.
NOTF	10 days later (H.P. Report: 10/5/55).
DIRC	Unannounced tour of the cyclotron. Visitors went from the exp. area to the cyc. pit through the open sliding shielding door (with safety interlock). The door was then closed by cyc. personnel and the beam turned on.
BEAM	Not specified.
REMK	Dose rate in occupied area estimates as 200 rads/hr.
TIME	Total of 25 sec: 10 sec in dir. beam area and 15 sec in lower level areas.
BODY	Assumed total body exposed to direct beam.
MONTR	None.
TESTS	Trial runs to determine the exposure time as given.
DOSE	From 1 to 5 rad per individual.
RECOM	Projects to improve safety devices.
DATE	1330 hours 10/7/1956.
PERS	Staff experimenter.
NOTF	Immediately after (H.P. Report: no date, but later than 10/10/56.)
CIRC	Working in exp. area while external beam was on. Inattention to standard precautions.
BEAM	External protons directed into a 6-ft lead cube.
REMK	Estimated dose rate in direct beam: 7.2×10^6 rads/hr. Fast neutron flux density in immediate areas around lead cube estimated in the range of 10^4 to 10^6 neutrons/cm^2 sec.
TIME	Max. of 8 min at minimum distance to dir. beam of 2 m.
BODY	Total body.
MONTR	β–γ film badge: 0.60 rem. Urine sample for slow neutron induced Na^{24} act.: 2 rads. Blood count: normal.
DOSE	From 0.60 to 2.0 rems.
RECOM	Temporary interlock system at exp. area entrances installed on 10/8/56.

Abbreviations used: DATE = Date; PERS = Personnel involved; NOTF = Notification delay to health physics; CIRC = Circumstances; BEAM = Accelerator beam characteristics; REMK = Remarks; TIME = Exposure time;

Table 4–(contd.)

DATE	1300 hours 2/4/1957.
PERS	Two workmen (Construction Co.).
NOTF	Immediately after (H.P. Preliminary Report: 2/7/51.
CIRC	Working in proton channel (for new Proton Station) with beam on (mesons to exp. area: Meson Station). Inattention to explicit H.P. recommendation for delay until after scheduled clearance survey.
BEAM	Clockwise (neutron spray toward Proton Station) protons on internal Be target. Intensity at ion-chamber monitor in cyc.-pit about 1.4×10^{-4} A.
REMK	(Announced H.P. Final Report not found).
TIME	Max. of 10 min.
BODY	Total body.
MONTR	Not specified.
TESTS	BF_3 in paraffin: 480 fast neutrons/cm² sec "Jordan" survey meter: 0.04 rad/hr (γ).
DOSE	Not specified.
RECOM	Not specified.
DATE	2230 hours 11/20/1957.
PERS	"Operator".
NOTF	1 hr 15 min later. (H.P. Report: no date).
CIRC	Submental 8 mm cut on chin while working in cyc.-pit.
BEAM	Off.
REMK	Admitted in emergency room of Univ. Clinics.
TIME	–
BODY	Chin.
MONTR	Not specified.
TESTS	Search for radioactive material in wound (with G.M. counter): negative.
DOSE	(Internal) Not detectable by means used.
RECOM	23 weeks away from radiation sources (First recommended 8/8/57 after the operators' film badge of 7/7/57 had shown a dose of 4.5 rads, with no evidence of invalidity).
DATE	1700 hours 2/16/1959.
PERS	"Operator".
NOTF	14 hr later (H.P. Preliminary Report: 2/17/59; Final Report: 2/24/59.
CIRC	Series of expeditious ion source alignments—with interlocks jumped—by operator running down (cyc.) and up (control rm.) while beam going off and on. After the fourth round trip, the unwanted combination was discovered: beam on, operator down.
BEAM	450-MeV protons on internal Be target. No. of prot/sec. on target: 2.3×10^{12}. Beam intensity on target: 0.4 μA. (measured target dissipated power: 8 W_i). Intensity at ion.-chamber monitor in cyc.-pit: about 3×10^{-6} A.
REMK	Estimated dose rate (RBE = 10) due to direct fast neutron spray, at 2 m from target, in median plane: 100 rems/sec per cm² of tissue.
TIME	Estimated by operator: 15 sec. Actual: 10.5 sec. At about 4 m from target, 10 in. below median plane.
BODY	Head and chest.
MONTR	2 pocket chambers: both off scale (0.2 r). β- + γ-film: 0.123 r. Thermal neutrons (Cd filter) 0.039 rem. (Assigned fast neutron film was not in badge.)
TESTS	Trial run exposing two sets of β- + γ- plus fast neutron films, and two pocket chambers. Actual exposure time calculated by comparison of incident and trial γ-films. Urine: no detectable amount of Na^{24} (medical analysis) Blood: no clinically important exposure (med. analysis). Eyes (8 months later): no radiation effect.
DOSE	γ: 0.123; fast neutron: 2.74; thermal neutron: 0.039; total: 2.902 rems.
RECOM	No restricted activities (based on : accumulated dose rate, incident included, 3.9 rems/year). Enforcement of standard safety procedures. Incitement to prompt notification to health physics.

BODY = Part of body exposed; MONTR = Personal monitors; TESTS = Additional tests; DOSE = Estimated dose; RECOM = Recommendations.

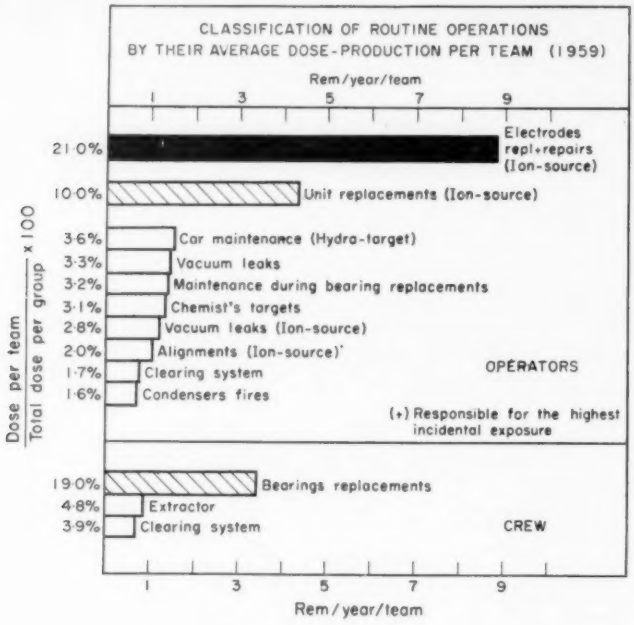

FIG. 7.

Non-routine operations

	(rem/man-year)
Wiring light-circuit in tank (1956)	1.80
Cleaning diffusion pumps (1957)	1.60
Shielding (1954, 1955, 1956)	0.98
Plastic windows (1956)	0.80
Hydra Top repairs (twice/3 years)	0.50
Dee cleaning (1957, 1959)	0.44

No. 4 *vacuum leaks*

Vacuum leaks have occurred about once a month on the average. The time required for detection and repair is exceedingly variable. Two hours is taken as a rough average. Maintenance is regularly performed on the vacuum seals of the rotary condenser along with bearing replacements (see cause no. 8). Two operators are usually required; they work in an area where the radiation level is approximately 0.03 r/hr.

No. 6 *target maintenance*

The "Hydra target" has been used since early in 1954. During its development period (from October 1953 to April 1959), two operators and one crew member entered the tank for 2 hr each, on three separate occasions. The radiation level within the tank at this time was about 0.2 r/hr.

Since then, maintenance has been performed on the unit about four times per year, major repairs being necessary about once each year. One or two operators work in a radiation level averaging 0.16 r/hr for about 1.25 hr during routine maintenance operations.

Majors repairs are dictated by failures in the car itself or in the top of the assembly. Car repair calls for one operator to remove and reinstall the unit (2.25 hr, average level 0.01 r/hr), and one machinist to disassemble and repair the car (2 hr, average level 0.01 r/hr, behind shield).

Failures in the top have occurred four times during the 6 years, and have been handled by one operator

working a maximum of 40 min inside the tank in a radiation level between 0.5 and 1.0 r/hr.

No. 7 *extractors*

The extractors, used to channel the external proton beam, were developed in 1955 along with the methods of polarizing, deflecting and focusing the beam.

Changes from meson to proton experiments and vice versa require, respectively, installation and removal of the extractors. With a view towards minimizing the number of these operations and the corresponding doses, the schedule shown in Table 1 has been adhered to. That is, one installation–removal cycle takes 16 weeks, the extractors being removed 7 weeks after installation. This corresponds to six or seven operations per year. Two crew members, one operator, and one research group member are required. The crew spends about 1 hr in a level of 0.07 r/hr; the operator (inside the tank), about 5 min at 1 r/hr, and the researcher 30 min in a level of 0.1 r/hr.

No. 8 *bearing replacements*

The two bearings for the shaft of the large rotary condenser have a life varying from 5 to 2500 hr. At present, they are usually replaced after two experiments (about every 6 weeks), if they have not previously failed. The time necessary for replacement has been reduced from 24 hr to 4 hr in 1959. The operation requires from four to six crew members and from one to three operators, and is performed in a level of 0.02 r/hr. Additional maintenance is often performed concurrently with the bearing replacements; e.g. ion source maintenance, vacuum leak detection, r.f. system inspection and adjustments, and water leak, flow and temperature checks.

No. 10 *dee cleaning*

Dee cleaning has been performed only two times since 1954, once in 1957 and again in 1959. A major operation, it necessitates the retraction of the entire radiofrequency system, and involves the cleaning of the following components: dee, dummy dee, dee stem, cathode loop parts, pole liners, stub and stub liners. The massive bulk of radioactive matter surrounding the working area (mostly copper activated by protons and neutrons for several years), the impracticability of a temporary shielding and the atmosphere of radioactive dust created by the wire brushing (to remove deposited oil film), make the procedure quite difficult and necessitate strict health physics supervision.

In 1957, an accidental oil spray made dee cleaning necessary. Thirteen operators, eight crew members, and eighteen research volunteers co-operated over a 2-week period, each man spending 20 min in a level of 1.2 r/hr.

In 1959, natural conditions demanded another cleaning. Wire brushing was done by ten volunteers (research) during 4-hr, each man working for 10 min. Eight crew members (10 min) and twelve operators (15 min) performed the other tasks. No doses exceeding 0.3 rem were recorded, although the level was as high as 1.8 r/hr.

No. 11 *stub cleaning*

Stub cleaning has been performed a total of six times, three times along with dee cleaning. The entire crew participates in the operation which is performed in a level averaging 0.02 r/hr, and is spread out over a 3-day period.

No. 12 *clearing system*

Removal of the clearing system is required for stub cleaning, shifting or replacement of the stators, and for shifting and/or replacement of the shorting bars. It has been performed a total of twelve times, each time with two operators to remove the external wires, and two crew members to remove the stub clearing skin. The procedure has taken about three days (10 working hours) and involves work in a level of 0.03 r/hr.

No. 13 *water cooling system*

Repair of water leaks in the vacuum tank is necessary about twice a year. The operation, which takes about 8 hr, involves draining the lines of water, detecting the leak with compressed air, and actual repair. One crew member works in a level of 0.02 r/hr.

Along with dee cleaning, the filter of the magnet coils is cleaned. This operation (draining and cleaning) occupies one or two crew members for an average of two days.

No. 14 *oil condenser fires*

Two to three times each year it has been necessary to rebuild the stub tank clearing wires because of fires caused by the oil condensers. Two operators work for 3–4 hr in a radiation level of about 0.04 r/hr.

No. 15 *diffusion pump maintenance*

Work on the diffusion pumps consists of disassembling and cleaning the pumps and/or repairing water leaks. Cleaning was performed in 1957, with two crew members working 24 hr at a level of 0.05 r/hr. In 1958 a water leak occurred which was repaired by two crew members in 1 week at 0.02 r/hr.

No. 17 *shielding*

Shielding improvements have been accomplished in six operations, three of which are of interest here.

In 1954, a supplement to the ion source shielding of 1953, was erected by from two to three crew members in 7 hr, in a level of 0.1 r/hr. The year 1955 saw the construction of the large rotary shielding. Construction took 20 days and required from four to six crew members, who spent a maximum of 8 hr in the tank vicinity (0.1 r/hr.). In 1956, it was necessary to cut slots in the 1953 ion source shielding in order to relocate the source. Two crew members completed the work in 30 hr, at 0.1 r/hr.

No. 18 *plastic windows*

In 1956, the plastic viewing windows in port no. 4 were replaced because of radiation damage. The work required two operators, one machinist, and one draftsman for about 8 hr, in a radiation level of 0.1 r/hr.

No. 19 *lights in tank*

Fluorescent lamps inside the tank have to be replaced at an average of twice a year. This requires one operator for about 3 min, at 1.0 r/hr.

In June 1956 the damaged electrical cables were replaced and the lighting system was improved. In the tank, two operators worked for 1 hr each, at 1.0 r/hr; by the terminal window two more operators spent 5 hr at 0.1 r/hr.

No. 20 2-in. *probe maintenance*

Probe maintenance occurs only infrequently. Probes are replaced about once every 1.5 years, by two operators. This operation takes 15 min at 0.1 r/hr. Air lock repairs are performed by two machinists, and require 5 hr work in a level of 0.1 r/hr, and are necessary only about once every 1.5 years.

In addition, the probe hole in port no. 3 was reamed in 1958 and 1959. Two machinists, working 12 hr at about 0.2 r/hr were required.

No. 21 *clipper changes*

A "flip" clipper, operating on a beam, no-beam basis, was originally installed and is still in place. In May 1956, a continuously adjustable clipper was installed by one physicist (16 hr), one machinist (8 hr), and two operators (8 hr each), at an estimated 0.2 r/hr.

Copper-lined parts in the tank are protected from vertical oscillations of the beam by two graphite blocks, installed in 1953 by two crew members in 1 hr at a level of 0.8 r/hr. Since then, these clippers have been removed and/or relocated several times for various reasons (installation of extractor, etc.). On these occasions, one operator, crew member or physicist has worked inside the tank (1.0 r/hr) for about 5 min.

No. 23 *radiochemistry*

Chemists expose targets to the beam about twice a week. The target in its holder is mounted on the 2-in. probe, which is manually positioned in the cyclotron by a chemist in a level of 0.2 r/hr, assisted by an operator in a level of 0.06 r/hr. This operation requires about 10 min.

Target removal takes no more than 3 min at about 0.3 r/hr for the chemist and 0.06 r/hr for the operator.

The activity of the holder-target unit (U, Al, Cu, etc.) averages 10 r/hr $\beta + \gamma$ and 0.6 r/hr γ at 25 cm, 5 min after bombardment.

ANL-Reactor targets have also been handled by the chemistry group at the rate of about nine per year. Gamma-activities were on the order of 1–3 r/hr, at 25 cm.

There are hundreds of other operations that require entrance into the cyclotron pit. Most of them either occur very infrequently or demand only a few minutes time; e.g. oiling of bearings, temperature and location checks, tube replacements, visitor tours, etc.

SUMMARY

(1) In this report, we have intended to give a detailed picture of the past operation of the 170-in. Synchrocyclotron, and to relate relevant circumstances to their consequences.

The compilations of data in Part I have the practical value of a record for future reference.

(2) More questionable, though, is the significance of the correlations presented in Part II, which result from a dubious interpretation of diverse and sometimes incomplete reports. The information given in "Data and Comments on Causes" must, however, serve as the cornerstone of most practical applications; e.g. dose estimates and work schedules. Accordingly, an overall estimate of its merit has been made by the following test.

(a) A table* with a summary of these data was prepared, in which assumed values were used where the original data was ambiguous or absent.

(b) This table was used to estimate the total dose per group for each of the past 6 years.

* EFINS Report No. 348.

(c) These estimated totals were compared with the actual (recorded) totals given in Table 2, of Part I. (Appropriate corrections for background were made).

The majority of the estimated doses fall within ± 10 per cent of the actual values, with a tendency to be overestimated. Two larger deviations are evident in the results for the "operator" group: 1954 (insufficient data), and 1958, an anomalous year, in which not a single overdose was recorded.

(3) Past experience shows that the actual capability of the Synchrocyclotron as a dose-producing machine is limited. The relatively effective system of shielding and interlocks, together with a few safety rules and the tendency to keep maintenance time at a minimum, combine to make "biological doses" very improbable. However, records also show that weekly, and occasionally quarterly, overdoses do occur; they were especially prevalent in the early years.

With the prospect of increasing output of the machine, shielding additions, and other developments (radiofrequency system, mesons in Proton Station, etc.), and the trend toward decreasing maximum permissible limits,* an improved dose administration is in order. Steps in this direction, particularly of an educational nature, have already been taken.

(4) This report would be incomplete without the mention of other factors, difficult to evaluate, but which undoubtedly contribute to overdoses.

The most important of these factors, in our opinion, are:

(a) Indifferent attitude of personnel toward dose-economy.

(b) Disregard for distributing doses equally among personnel, in planning work.

(c) Underestimation of importance of duration and frequency of work in relatively "cool" areas.

The influence of these factors can be realized by comparing the years 1956–1957 with a similar maintenance period, 1958–1959, during which time an intensive attempt was made to minimize them.

* Last restriction effective January 1961.

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SEA WATER MONITORING FOLLOWING RADIOACTIVE WASTE DISPOSAL OPERATIONS

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Abstract—The growing concern of state and local regulatory agencies with regard to the disposal of radioactive waste is emphasizing the importance of environmental monitoring data. Although such data are routinely obtained for inland disposal operations, monitoring of sea disposal areas by the disposing agencies has received little attention. This article describes some of the problems associated with sea water monitoring, and presents analytical procedures that have been used at Atomics International for sample analysis and data interpretation.

INTRODUCTION

THE expansion of the nuclear industry in the United States during recent years has created a great deal of interest among state and local regulatory agencies with regard to the disposal of radioactive wastes. Some agencies are already investigating disposal problems and taking positive action toward control programs. For example, the California State Health Department, and the California State Department of Fish and Game, recently petitioned the Federal Government to limit the disposal of radioactive wastes at sea to a minimum depth of 2000 fathoms,[1] rather than the 1000 fathoms limit recommended by the National Committee on Radiation Protection.[2] Actions of this nature may be expected to affect disposal programs at many nuclear facilities, and the need for liaison between facilities and agencies is clearly indicated if unnecessarily severe restrictions are to be avoided.

Environmental monitoring has been one of the most successful procedures for controlling hazards which might result from the disposal of radioactive wastes. Samples are obtained from the environment of the disposed material and analyzed for radioactive isotopes. These environments, water, soil, air, vegetation, etc., always contain naturally occurring radioactive elements and usually contain additional radioactive materials due to nuclear weapons test fallout. It is necessary to compare the results from waste environment samples with results from identical background samples obtained before or during deposition of the waste in order to determine the extent of environmental contamination caused by the waste. If the circumstances of the disposal operation allow concurrent sampling from the waste and background environments, the important problem of variable fallout concentrations is minimized. Unfortunately, such is usually not the case, and the chronology of nuclear weapons testing must be considered when comparing sample results with background samples obtained months or even years previously.

Despite limitations, environmental monitoring results can play an important role in facility-agency relations where radioactive waste disposal is concerned. If maintained open to agency inspection, they are indicative of the desire to maintain a safe operation, of the presence of adequate technical competence, and they usually afford conclusive evidence as to whether a hazard has been created.

Of the presently accepted disposal techniques, sea disposal presents the most difficult challenge to environmental monitoring. The very features which make the ocean an attractive

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disposal area, also account for several monitoring perplexities: (1) the enormous volume of the ocean provides excellent dilution, but masks the migration of radioactive materials that escape their containers; (2) the great ocean depths provide ideal inaccessibility, but hinder adequate depth sampling; (3) due to its depth, the water which receives radioactive waste is believed to be of limited significance with respect to man's food chain, but this depth complicates any effort to secure marine life specimens for ecological cycle studies; (4) the composition of sea water limits its usefulness to man, but creates several problems for the radiochemist who must analyze the samples. Although it is not easy to evaluate the hazards which may be associated with sea disposal, the sea continues to be a popular disposal area, and there are indications that low level radioactive waste will be disposed in this manner for some time to come.[3] The need for environmental monitoring, conducted by the disposer, is perhaps as real in this area of waste disposal as any other.

MONITORING TECHNIQUES

The monitoring technique employed must be compatible with the funds available as determined by the expected value of the effort. This is, at present, an arbitrary decision because sea water monitoring is not required in conjunction with disposal operations. A limited program designed to provide data for review by regulatory agencies can be justified. Such data, of course, would be considered as supplementary to the rather exhaustive general studies which are now being performed for the United States AEC.[3,4] Most of the wastes that are approved for sea disposal are of very low curiage,[3–5] so that direct monitoring with submergable detectors does not provide sufficient sensitivity.[6] Sample analysis is a more logical approach for a routine monitoring program.

Sample collection procedures are quite flexible, and are dependent largely on the purpose of the program. Two examples may serve to illustrate this point.

The practice of bulk disposal, i.e. the direct release to the ocean surface of liquid radioactive waste, is not widely accepted in this country today. In like manner, the sea disposal of grossly contaminated equipment, without packaging provisions adequate to prevent surface water contamination, is not considered acceptable. It is believed that such practices, unless they are carefully controlled, can result in an undesirable concentration of radioactive materials in edible marine life. If a disposer desires to obtain data indicating that he does not perform such operations, or that his operations of this type are safe, samples taken from the surface may serve this purpose. An additional sample, for background reference, should be obtained a few miles from the disposal area. The problem of variable fallout is minimized in this manner, provided that the time lag between samples is small, and that rainout has not distorted the picture.

Most of the radioactive waste disposed to the sea is contained in packages designed to prevent premature releases of contaminants and to prevent the escape of waste items that could float.[2,7] If a disposer desires to obtain data indicating the adequacy of his containment design, depth samples are indicated. Such samples are obtainable with a Nansen bottle, Bacon Bomb Sampler, or similar depth sampling apparatus.[8] Mid-depth samples, before and after, are indicative of pressure damage to the containers during descent, while bottom samples may indicate the degree of contaminant release from containers disposed previously, or the manner in which present containers withstood the crash on to the ocean floor.[9] It is noteworthy that oceanographers who are studying these problems have some evidence that dispersal on the ocean floor may be preferable to attempted total containment.*

RADIOANALYSIS PROBLEMS

Samples should be analyzed for the most likely contaminants. Isotopic, rather than gross

* Dr. J. D. Isaacs, of Scripps Institution of Oceanography, in the paper, "Fundamentals of Dispersion and Diffusion in the Ocean", presented in Boston, Mass., at the 1960 Annual Health Physics Society Meeting, indicated that attempts at total containment could eventually result in the concentration of radioisotopes in marine life forms which attach themselves to containers and which later become food for more mobile marine life.

Table 1. Naturally occurring radioisotopes in sea water

Nuclide[10]	Concentration[10] (μc/ml)	β-Decay of principle contributors[21]
K^{40}	3.24×10^{-7}	89%, 1.33 MeV (max.)
Rb^{87}	5.94×10^{-9}	100%, 0.27 MeV (max.)
U^{238}	2.7×10^{-9}†	100%, 0.13(Th^{234}) and 2.32(Pa^{234}) MeV (max.), to Ra^{226}
Ra^{226}	8.1×10^{-10}†	~100%, 0.65(Pb^{214}), 3.15(Bi^{214}), 0.025(Pb^{210}), 1.17(Bi^{210}), 1.51(Tl^{206}) MeV (max.)
U^{235}	8.1×10^{-11}†	negligible
Th^{232}	5.4×10^{-12}†	negligible
C^{14}	1.9×10^{-10}	100%, 0.158 MeV (max.)
H^{3}‡	6.8×10^{-10}	100%, 0.018 MeV (max.).

† Activity of nuclide + daughter products.
‡ Only in top 50–100 meters of the ocean.

activity, analysis is dictated by the high salts concentration of sea water. If simple evaporation is attempted, the residue from untreated sea water is so great that it is impossible to compact the quantity required for a statistically significant counting rate into a counting planchet.

The naturally occurring radioisotopes in sea water are listed in Table 1, from the data of Revelle *et al.*[10] It is apparent that the high potassium content of sea water could easily result in misleading counting data interpretations, because variations in potassium concentration between background and disposal area samples could indicate β-emitting contaminants. Donaldson *et al.*[11] and Harley[12] have reported the K^{40} isotope activity in sea water as varying from 600–700 dis/min per l., whereas sample variations much smaller may be of interest. Since the K^{40} β-contribution is almost two orders of magnitude greater than the usual mixed fission product contribution (cf. Table 2), a chemical separation procedure with very low potassium recovery is required. Of the other nuclides present, only Rb^{87} and a few of the U^{238} daughters are of sufficient abundance and

Table 2. Surface water monitoring results for seven sea disposal operations

Date	Type	Mixed fission products (μc/ml $\times 10^{-9}$)	Uranium (μg/l.)
October 1957	Background	22.0	—
	Area	20.0	—
May 1958	Background	13.4	5
	Area	28.8	10
July 1958	Background	7.5	15
	Area	8.1	8
September 1958	Background	7.5	5
	Area	1.6	5
March 1959	Background	5.0	2
	Area	2.5	2
April 1959	Background	3.0	1
	Area	5.2	2
June 1959	Background	10.8	8
	Area	7.8	3

β-energy to affect counting rates appreciably. Even if these nuclides were totally recovered in the sample preparation process, their contribution would be too small to mask the fission products and would be similar in both background and area samples. The lower fission product values in Table 2 suggest that these nuclides are not recovered efficiently by the chemical procedure that was used.

The presence of plankton in the ocean is variable, and plankton are efficient concentrating agents for radioactive material.[13] Radioactivity concentrations in plankton have been reported as varying between 1.3×10^3 and 1.1×10^6 dis/min per g, with average values 7.1×10^3 times the average surface water concentration.[11] As with K^{40}, a very significant concentration differential between background and area samples can result. This problem is intensified by the high efficiency of precipitation procedures for plankton removal. Pre-filtering of samples to remove plankton is unwise, because waste contaminants could be in insoluble form.

Fission product age can be an analytical problem if a gross fission product precipitation procedure is used. The isotopic distribution of mixed fission products is changed considerably as the short-lived isotopes decay, producing important effects on the relative contribution to total β-activity (cf. Fig. 2). If there is a large age difference between actual sample fission products and those used in the laboratory to determine the precipitation efficiency, a misleading result may be obtained. To illustrate, a given precipitation procedure may be very specific for a relatively short-lived isotope. If fresh fission-product spikes are used to find the recovery efficiency of the procedure, a rather high yield may be expected. Should this yield factor then be used to calculate the concentration in samples contaminated by aged fission products, a low value would be obtained.

Any environmental monitoring effort of this nature is complicated by the presence of nuclear weapons testing debris. The data reported in Table 2, when considered in the light of weapons testing chronology, reveals that fallout near the Pacific Coast has trebled surface water concentrations in the past. The fallout error is uncontrollable, and virtually impossible to deal with statistically. However, it is believed that natural movements of the sea produce a degree of homogeniety which increases with depth. Since the disposal area samples are compared with background samples obtained under similar conditions, the fallout error is small unless the sampler has the bad luck of taking his samples during unfavorable meteorological conditions that distort his results.

A final problem is created by the statistics of sample comparison. The maximum contamination level that might reasonably be expected from disposal operations is quite low, possibly below detection limits. The monitoring procedure must therefore, of necessity, be designed to obtain valid negative data. Such data are of value if intended to show the absence of hazard, provided that good sampling and testing procedures can be demonstrated. The large counting errors that accompany low-level analysis intensify this problem. Recognition of these considerations suggests that a realistic differential should be selected between background and area samples, above which sample variations are considered to be significant. Ideally, this differential would rule out natural sample variations as insignificant, with an acceptable degree of confidence.

ANALYTICAL PROCEDURES

Although sea water is not easily analyzed for radioactive material, two relatively inexpensive procedures are suggested below for two common contaminants—uranium and gross fission products. These procedures were used successfully at Atomics International for seawater monitoring purposes.

A polarographic technique was used for the uranium analysis.[14] One liter of sea water is evaporated in acid solution. The uranium is complexed as the diethyldithiocarbamate, extracted into chloroform, and analyzed with a polarographic apparatus. Uranium is identified by a characteristic voltage value at its valence reduction. The quantity is measured by the current flow increase at this characteristic voltage. This procedure is accurate to ± 1 μg. The average uranium concentration in the ocean is about 3 μg/l., and a 1-l. sample provides normal background accuracy of ± 33 per cent.[15]

A co-precipitation technique was selected for mixed fission products to avoid the high salts concentration and K^{40} problems. $CaCO_3$ is precipitated

from hot 250-ml samples by adding 5 ml 3 N Na_2CO_3 solution. With the exception of $CsBa^{137}$, which is retained in the filtrate, and non-volatile $RuRh^{106}$ and Nb^{95} which are partially retained, aged fission products are co-precipitated with the calcium.[11,12] Using a flame photometric procedure, it was found that the potassium recovery averages only 1.5 per cent.[16] A millipore filter system is used to collect the precipitate, which is ashed at 350°C in a muffle furnace. The sample solutions are maintained at near 100°C during the filtering process to prevent the return of $CaCO_3$ to solution.[17] The final residue is weighed in stainless steel planchets and counted in a proportional system.

Other workers using this technique have reported co-precipitation efficiency for aged fission products as high as 77 per cent.[1] Since the recovery factor is

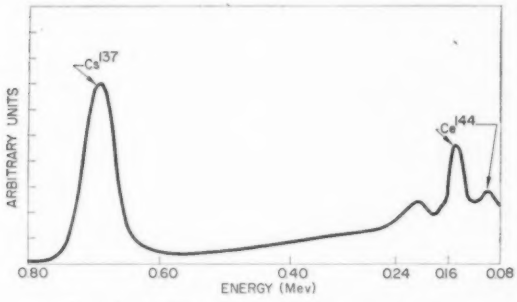

FIG. 1. MFP source solution γ-spectrum.

important in the determination of the fission product concentration, a series of control samples were spiked with mixed fission products and processed to obtain verification. The following procedure was used to minimize errors due to self-absorption, geometry and scattering. An aliquot of the contaminant was added to each sea water sample, and the entire procedure performed. A weight of NaCl, identical to the total ash weight of each sample, was dissolved in distilled water, and an identical aliquot of the contaminant added. These solutions were slowly evaporated, transferred to planchets, and counted. Assuming homogeneity in each sample, the self-absorption, geometry, and scattering were virtually the same, and the count rate ratios correspond to the fractional recovery. An average recovery of only 71 per cent was obtained. A γ-spectrum of the spike, shown in Fig. 1, revealed the Ce^{144} γ's to be a small fraction of the $CsBa^{137}$ γ's, indicating a source age of several years.[18] It was decided that dissimilar spike ages were the most likely cause of the gross recovery difference, due to preferential co-precipitation of shorter-lived nuclides. Thus a question was raised as

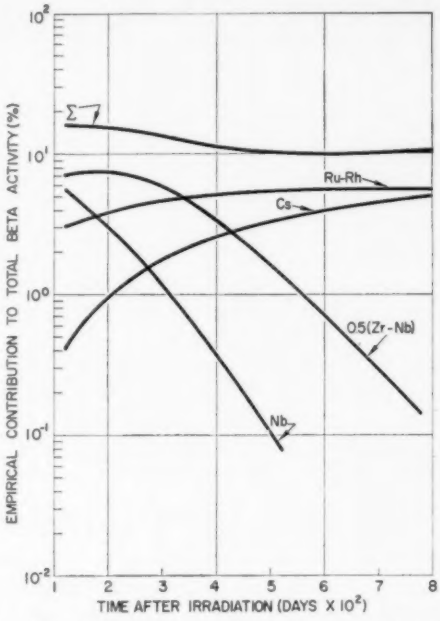

FIG. 2. Recovery variation due to MFP age.

to the magnitude of error which could result in the monitoring program from age differences between the spike and actual sample contaminants.

The error magnitude was evaluated in a simple manner, as shown in Fig. 2, using the data of RUBINSON *et al.*[18] Here the relative β-contributions from those major isotopes which are not recovered efficiently are summed as a function of time. The essentially zero slope for the summation, after 500 days, indicates negligible error for fission products of the age normally disposed. It is assumed that the slope is not affected by the partial nature of Nb and RuRh recovery.

Figure 3 shows the β gross counting efficiency curve

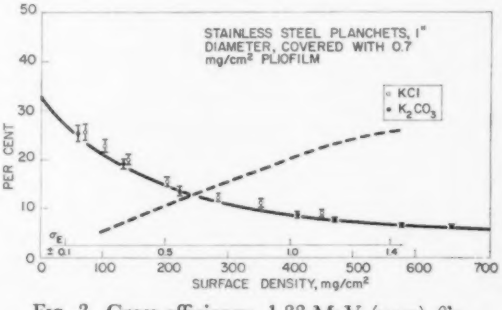

FIG. 3. Gross efficiency, 1.33 MeV (max) β's.

obtained with graduated quantities of two potassium compounds, KCl and K_2CO_3. The K^{40} isotope undergoes β-decay of 1.33 MeV (max.), which approximates the effective energy of aged mixed fission products, and has an easily determined specific activity in several readily available compounds. For these reasons it was chosen as the standard, and the results reported in Table 2 are K^{40} equivalent. Figure 3 shows that, due to low specific activity, an acceptable standard deviation, σ_x cannot be achieved at a gross efficiency above 25 per cent. The "zero" absorber point was obtained with a very thin aliquot of this spike.

The mixed fission product concentration for a given sample is given by

$$x = \frac{r \times 4.5 \times 10^{-7}}{0.71\ VE} \tag{1}$$

where x = concentration (μc/ml);
r = net count rate (counts/min);
0.71 = co-precipitation efficiency;
V = sample volume (ml);
E = gross counting efficiency.

DATA INTERPRETATION

For the mixed fission product analysis, a negative or positive differential will usually exist between the area and background samples due to expected variations in fission-product recovery, fallout, counting, plankton, gross efficiency, or some combination of these. A statistical test for this differential may be selected such that the presence of an extraneous contaminant (such as from radioactive waste) is indicated only when the background and area samples satisfy the test, or a simple limit such as twice background may be used. From Table 2, it is apparent that routine results will often vary by a factor of two or more.

A rigorous test for the difference between two means has been derived from the "Student" t-distribution.[19] This distribution is very similar to the normal distribution, except that greater probabilities of large deviations from the mean are involved for small groups. Application of this test here is essentially a determination as to whether two independent samples, area and background, came from different normally distributed populations, viz. natural sea water and sea water contaminated by radioactive waste. The quantity t is first determined as:

$$t = \frac{[(\bar{x}_1 - \bar{x}_2)]\sqrt{[(n_1 + n_2 - 2)n_1n_2]}}{\sqrt{[(n_1 + n_2)(n_1s_1^2 + n_2s_2^2)]}} \tag{2}$$

where $\bar{x}_1$ and $\bar{x}_2$ are the means of the two samples (in this case in units of microcuries per milliliter $\times 10^{-9}$), n_1 and n_2 designate the number of repetitions of the analytical procedure performed for each sample, and s_1^2 and s_2^2 designate the corresponding sample variances, s being the standard deviation. Here s must be computed by combining the standard deviation σ_1 obtained from n procedures

$$\sigma_1^2 = \sum_{i=1}^{n} (x_i - \bar{x})^2/n \tag{3}$$

with the mean counting error σ_2, which may be much larger.

$$\sigma_2^2 = \sum_{i=1}^{n} \left(\frac{N_i}{T_i} + \frac{N^*}{T^*}\right)/n \tag{4}$$

where N = sample gross count rate (counts/min);
T = sample counting time (min);
N^* = counter background count rate (counts/min);
T^* = background counting time (min).

If σ_1 is considered as the error of the analytical technique, and σ_2 regarded as the error due to the random nature of radioactive decay, the two are independent, and may be combined as the square root of the sum of the squares,[20] therefore

$$s^2 = \sigma_1^2 + \sigma_2^2 \tag{5}$$

Table 3 presents values of t_ε for several levels of significance ε, according to the number of degrees of freedom m,

$$m = n_1 + n_2 - 2 \tag{6}$$

The level of significance may be selected arbitrarily, according to the desired degree of certainty as to whether the samples did come from different populations. For a given ε, if t, as calculated from (2), is greater than t_ε, obtained from Table 3, then the probability that the samples came from different populations is at least $1 - \varepsilon$. When the area sample concentration exceeds that of the background sample

Table 3. Values of t_ε for the indicated degrees of freedom and level of significance[19]

m	$\varepsilon = 0.10$	$\varepsilon = 0.05$	$\varepsilon = 0.01$
1	6.314	12.71	63.66
2	2.920	4.303	9.925
3	2.353	3.182	5.841
4	2.132	2.776	4.604
5	2.015	2.571	4.032
6	1.943	2.447	3.707
7	1.895	2.365	3.499
8	1.860	2.306	3.355
9	1.833	2.262	3.250
10	1.812	2.228	3.169

sufficiently to provide a high probability that the samples are from different populations, there is still no absolute basis for a decision as to whether the difference resulted from waste disposal operations or from other variables. However, the purpose of the procedure is to provide reliable negative information.

Table 4 presents an example application of this procedure for data interpretation. Four complete procedures are performed for each sample, the results averaged, and the variations computed from equations (3), (4) and (5). The magnitude of t is calculated from equation (2) as 2.0, and compared with $t_\varepsilon = 2.447$ for the 0.05 significance level at $m = 6$. Since $t < t_\varepsilon$, the data indicate that there is less than 95 per cent probability that the samples came from different populations. A statistical method of this nature provides little certainty that both samples came from sea water of identical radioactive material content. Also, an evaluation of the significance of any detected increase over background is determined from the magnitude of the net area sample concentration, rather than from a statistical analysis. The statistical method only indicates whether the difference between two means is sufficiently large, as compared with deviations in individual radioanalyses from each sample, to justify concern about the magnitude of the difference, provided that the number of individual radioanalyses is acceptable.

Table 4. Example applications of "Student" t-test to sea water monitoring results

	Background sample	Area sample
x_i	9.9, 21.7, 10.2, 11.8	34.6, 25.4, 19.2, 36.0
$\bar{x}$	13.4	28.0
n	4	4
σ_1	4.8	6.9
σ_2	7.6	7.2
S^2	81.2	99.1
m	6	6
t	2.0	2.0
t_ε	2.447	2.447

CONCLUSION

Clearly, what is needed, to enable evaluation of the significance of the results of the statistical procedure, is the establishment of a sampling method and a value of ε such that, if the results fail to satisfy the "Student" t-test, there should be no concern about contamination of the disposal area water. These procedures should, preferably, be established on the basis of a detailed study of variations in background radioactivity levels and permissible contamination levels in sea water.

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NOTES

Health Physics for the N.S. *Savannah**†

(*Received* 1 *September* 1960)

Introduction

HEALTH physics considerations for the 20,000-ton nuclear ship *Savannah* have been an important part of the over-all design, construction and operational plans of the Government, and of States Marine Lines, Inc., who will operate the ship as the General Agent for the Maritime Administration. The philosophy that the *Savannah* would be one of the safest ships afloat, in addition to satisfying domestic and international radiation safety requirements, has led to maximum efforts in developing the applied health physics programs for the *Savannah*.

Savannah program

It is appropriate to point out that the health physics program for the operation of the ship, the maintenance of the 69-MW pressurized water reactor, and the protection of the ship's environment represent the consolidated efforts of the Atomic Energy Commission, the Maritime Administration, Oak Ridge National Laboratory and States Marine Lines, Inc. as General Agents for the U.S. Maritime Administration. Health physics programs at many reactor sites were examined in detail for features and techniques which would be applicable to a shipboard radiological protection program. In addition, operational experience of the nuclear submarine has been integrated into the development of the *Savannah* health physics program.

As a prototype, or so-called "first generation" nuclear merchant ship, the *Savannah* will have a complete health physics staff to conduct the comprehensive program planned for the ship. Such an elaborate program, however, is not anticipated for a commercially competitive vessel since it is expected that the operating experience gained during the start-up, testing and precommercial service periods of the *Savannah* will provide a baseline for the health physics requirements for future generations of commercial nuclear ships.

In the interest of safety and to insure its self-sufficiency in health physics while away from home port, radiological facilities on the *Savannah* have required extensive planning for all conceivable conditions, both nautical and nuclear.

During start-up and early trials, the health physics staff will be comprised of a senior health physicist on the technical staff of the General Agent, three technical assistants on loan from facilities of the Atomic Energy Commission and the Public Health Service. Three of the specially trained *Savannah* engineering officers will round out the health physics staff during the pre-commercial periods. Additionally, the ship's Medical Officer is a well-trained specialist in the field of radiation protection and industrial medicine. The health physics staff which will remain aboard for routine operation following the pre-commercial period will be specified as a result of experience gained during the initial phases of ship operation.

Exposure control

It is anticipated that radiation exposure to operating personnel will be well below the annual 5-rem occupational dose limit and that the annual dose for passengers will be below 500 mrems. To insure the integrity of the shielding, a special shield survey will be conducted by a team of specialists from the Oak Ridge National Laboratory during the test and trials program.

The ship will conduct and maintain an extensive bioassay program under the direction of the Medical Officer. The physical condition of all personnel serving aboard the ship will be screened periodically in addition to the bioassay services.

Shipboard facilities

The *Savannah* is outfitted with a complete health physics laboratory designed along lines generally similar to those aboard nuclear submarines. With substantially more available space, however, the laboratory is considerably more elaborate and will permit the use of more-refined analytical procedures and supplementary services. To implement the over-all program, a variety of versatile equipment

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† This paper is a resume of the shipboard, shipyard and enviromental aspects of radiological health as developed and applied to the nuclear merchant ship program, specifically for the passenger–cargo ship Savannah. A more complete summary is available on request from the Office of Research and Development, Maritime Administration.

has been provided in the laboratory, including a G–M detector, an α-scintillation unit, and a multichannel γ-spectrometer.

For personnel dosimetry, recommendations of the Health Physics Division of Oak Ridge National Laboratory have been followed to utilize the features of the Oak Ridge National Laboratory Film Badge Meter No. 2, complete with neutron foils in addition to Hurst Threshold Detectors. Equipment for a complete film badge program is aboard, including calibration facilities for badges as well as for the fixed and portable survey instruments.

These measures were deemed essential in meeting the design philosophy of self-sufficiency under all conditions, anywhere in the world.

In addition to the shipboard program, a practical environmental survey program has been developed on the basis of analytical techniques employed by the Health and Safety Laboratory of the Atomic Energy Commission, Oak Ridge National Laboratory, and the Public Health Service. It is planned that the *Savannah* will collect representative samples in each port area it visits. Duplicate sampling techniques will be employed in order that shipboard analytical results may be cross-checked with results obtained by port area or Government laboratories.

Monitoring system

In addition to facilities and portable equipment of the health physics program, the *Savannah* is equipped with a complete system of automatic radiation monitoring instruments (RMS) to measure and record area dose rates, air particulate and radio-gas levels, primary system activities, and associated engineering information. To back up this system, an extensive area survey will provide verification of measurements obtained from the fixed health physics monitoring systems. These calibration checks will be conducted routinely until operational experience allows reliance on the automatic systems with routine surveys.

Protective measures

Since the *Savannah* has been designed to operate safely under a wide range of conditions, an extensive personnel air-line breathing system has been designed and installed within the containment vessel and in the undershield demineralizer area to permit limited access during shut-down even in the presence of airborne fission product activity above MPC levels. This system will provide filtered air, monitored for carbon monoxide, to manifold outlets in the containment vessel and demineralizer space. Several types of respiratory equipment have been especially designed for this installation to provide versatility under any circumstances.

Sufficient protective clothing will be on the *Savannah* and properly stored to meet any emergency situation. Laundering of this apparel will be provided by contractural services at port facilities.

Access control

The *Savannah*, as in land-based reactor plants, will have certain areas to which personnel access is denied except under strict health physics supervision. Although during routine inspection and maintenance this should pose no major problem, it is necessary to consider that there could exist certain emergency conditions in which immediate access to a controlled area is mandatory. In planning for these situations, emergency procedures have been established in which health physics personnel will make simultaneous entry into controlled areas with Engine Department personnel in order to monitor these operations closely, and thus minimize exposure.

Waste management

In the absence of international policies for the disposal of radioactive wastes at sea, the *Savannah* has been designed with complete systems for collection and retention of radioactive wastes. It is planned that the ship will discharge neither liquid nor solid radioactive wastes to the environment. These wastes, including expended ion exchange resins, will be transferred periodically to the nuclear servicing vessel or to a licensed AEC contractor for processing and disposal. The release of airborne particulate and gaseous activities will be reduced by the use of filters and gas-adsorption units, or completely retained by discharge into the containment vessel, until such times as meteorological conditions are suitable for the controlled release of these materials.

Servicing

Servicing of mobile nuclear power plants has introduced unique factors into routine ship maintenance and overhaul procedures. In considering the feasibility of servicing facilities for shipboard reactors, several types were compared for convenience, safety and flexibility of servicing operations, in view of the potential numbers of nuclear ships. For support of the *Savannah* as well as other nuclear ships, a specially designed, 129-ft barge, the Nuclear Servicing Vessel *Atomic Servant*, has been constructed as a mobile facility for major reactor system maintenance, refueling operations and processing of reactor wastes.

Based upon preliminary design by the Electric Boat Division of General Dynamics, the *Atomic Servant* was launched earlier this month by Todd Shipyards, at Houston, Texas, for the Maritime Administration.

Waste-processing equipment and a spent-fuel-element storage pit are located below the main deck, while above it are areas for personnel decontamination, laboratory, laundry and a combination decontamination-workshop area as well as protective-clothing and equipment storage and issue rooms. The Operations Center of the *Atomic Servant* is located in the after end of the deck house. There will not be crew's quarters aboard since the vessel is not self-propelled and will be used principally in conjunction with conventional dockside facilities.

During refueling operations, a heavy-duty marine or shipyard crane will be used to lift and remove the upper head of the reactor pressure vessel and move the specially designed telescopic fuel-element transfer cask from the barge to the *Savannah*. Spent fuel elements will be stored under water in the shielded fuel-storage pit on the *Atomic Servant* for approximately ninety days before shipment to ORNL for reprocessing. Visual inspection of stored components is provided through a lead glass window located on one side of the shielded fuel pit. Defective fuel elements will be sealed within special canisters before placement in the transportation casks.

Nuclear-waste process systems provided on the barge will receive, process and package for disposal of radioactive solid and liquid wastes generated by the *Savannah*. Transfer of liquids and flushed ion-exchange resins from the *Savannah* will be through jacketed or doubly contained hoses. Activity of the low-level liquid wastes from the reactor primary coolant and primary shield water will be concentrated by demineralizers until the effluent activity has been reduced to acceptable levels for overboard discharge. Depending, however, on location at the time, these effluents may be retained aboard in storage tanks. Discharge of low-level liquids to the environment will be controlled by a locked check valve on the pump discharge close to the point of discharge and monitored for significant activity.

High-level wastes such as spent ion-exchange resins from the purification systems of the ship and barge will be packaged in pre-cast concrete forms. Other solid wastes, such as expanded charcoal adsorption units, as well as other solid items which cannot be decontaminated, will also be encased in concrete for ultimate land or sea disposal.

Careful design and control of exhaust systems on the barge will preclude problems of airborne contamination. These exhausts will be vented through particulate filters in the discharge stack and the effluent will be monitored for particulate and gaseous activity. In the event that radiation levels above acceptable values for release to the atmosphere are detected, the ventilation intakes will be secured and the ratio of diluting air will be increased correspondingly.

Instrumentation installed at selected working locations on the *Atomic Servant* will provide continuous monitoring of ambient radiation, air activity levels, and liquid-waste-effluent activity. Fluctuations above pre-set levels will activate alarms locally and in the health physics laboratory. Immediate surveys will be made and evaluated according to the hazards present, and corrective or preventive measures will be taken.

A complete system of records for reactor servicing, waste processing and disposal, as well as the more conventional accounting of presonnel exposure, will be maintained aboard the *Atomic Servant* and provided, as required, for periodic review.

Pre-operational survey

At the *Savannah* construction site in Camden, New Jersey, a plan for radiation surveillance in the Delaware River Valley was developed on the basis of the type of proposed nuclear operations, and on the unique local characteristics of the site. The surrounding area, having a relatively high population density as well as important agricultural areas, required that a wide variety of samples be collected.

An extensive research-type environmental sampling program has been conducted since 1958 through the co-operative efforts of state and local agencies of Pennsylvania, New Jersey and Delaware. The radiochemical analyses of field samples was performed by the U.S. Public Health Service at their Cincinnati laboratories. In addition to off-site surveillance, the New York Shipbuilding Corporation has conducted an on-site sampling program of air, water and surface smears. Air has been sampled at two stations within the shipyard for particulate activity. Water samples have been taken routinely from the Delaware River, shipyard wells and the municipal supplies of Camden and Gloucester, serving different areas in the yard. Surface smears are taken from ten representative locations within the yard.

Off-site samples have been collected periodically at seven stations in the Delaware River from Trenton, N.J. to the lower Delaware Bay. These have been a monthly composite with the exception of the Schuylkill River sample, which was initially collected on a weekly basis in an attempt to detect iodine-131 possibly discharged from medical research laboratories in the area. Results were negative, and sampling in the Schuylkill was changed to a monthly basis. Silt and algae samples are also collected at these seven locations, as available. Algae and shellfish are collected at the two Delaware Bay sampling stations.

Separate or special samples of airborne particulate

activity have not been collected, since the area is adequately covered by the National Air Sampling and Radiation Surveillance Networks of the Public Health Srevice. Precipitation samples are collected at five locations generally surrounding the site in New Jersey and Pennsylvania.

Agricultural samples have included five representative types of vegetables and two types of fruits as well as soil and fertilizer samples from the same fields at five separate locations. These specimens selected include asparagus (stem), cabbage (leaf), tomatoes (fruit), corn (grain), sweet potatoes (tuber) and apples and peaches (fruit). Milk samples have been taken from two locations, one each in Pennsylvania and New Jersey, with analysis by γ-spectrum. Analysis has included gross β on most samples, strontium-90 on selected samples and principally γ-spectrum of crop samples.

To determine existence of iodine-131 contamination, wild-animal thyroids (raccoons) were collected and submitted for analysis. Since weapons tests were suspended in November 1958, no evidence of iodine was found in any of the thyroid samples analyzed.

Examination of the theoretical consequences of the maximum credible accident

An essential part of every reactor safeguards evaluation is the analysis of the theoretical consequences of reactor accidents on the surrounding environment. This type of analysis for the *Savannah* was conducted by Oak Ridge National Laboratory. Selected portions of this study concerned with the behavior of radioactive contaminants in a tidal estuary are partially covered PRITCHARD[1] and PARKER *et al.*[2] Because of the metropolitan development of most port areas, this analysis has covered a wide variety of hypothesized accidents which might result in the release of radioactivity to the environment. However, of these, none are seen to result in exposures in excess of those considered permissible.

This basic analysis is being used by the Office of Research and Development of the Maritime Administration in development of operational plans for the *Savannah*'s ports of call and servicing sites. Although refueling sites have not been finally determined at this time, environmental evaluation will play an important part in their selection.

Summary

The shipboard design of Health Physics facilities and operational planning for the Nuclear Ship *Savannah* have been predicted on providing the highest degree of nuclear and nautical safety for passengers, crew and environment under all conceivable conditions. This planning and design for containment, shielding, monitoring and waste management will provide a unique baseline of operating experience which will then be applied in the design of future economically competitive nuclear merchant ships.

E. P. RESNER

State Marine Lines, Inc.
New York Shipbuilding Corporation
Camden, New Jersey

G. D. SCHMIDT
R. J. VAN TURNER

U.S. Public Health Service
assigned to Maritime
Administration

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Development of Safety Performance Tests for Radioisotope Sealed Sources and Devices*

(*Received* 17 *June* 1961)

Introduction

THE benefits available through radioisotopes were a subject of considerable interest to medical and scientific research workers prior to World War II. However, isotopes were limited in supply and they were very costly because they were produced only by cyclotrons and accelerators. With the advent of the nuclear reactor and its almost unlimited capacity for massive, low-cost production of isotopes, the door opened wide to extensive utilization of these unique tools, not only in science and medicine, but in almost every facet of U.S. industry.

These by-product materials of the atomic energy program have emerged as a major, if not the most important, tool or product of atomic energy. Shipments of over a million curies to date from Oak Ridge, plus almost an additional $\frac{3}{4}$ million curies imported and an estimated U.S. market of 3.5 million curies during the next two years, attest to their importance. The major fraction of these curie quantities has been used as sealed sources, ranging from a few microcuries as radiation standards to multi-kilocurie levels for large γ-irradiators. For the most part, sealed-source uses involve in-between quantities, from millicuries to kilocuries, for industrial

* Presented by James W. Hitch at the Health Physics Meeting, Las Vegas, Nevada, June 13–16, 1961.

gaging, radiography, static control, medical teletherapy and many other useful applications.

Isotopes are not unlike other useful tools or materials in that, to obtain their benefits, one must also accept some risks. We are all well aware of the many dangers as well as benefits associated with the automobile, the airplane, electricity, fire and even water itself. Yet, few of us would willingly forego the everyday benefits available through these media. Fortunately, the developers of atomic energy recognized very early that hazards are associated with the utilization of atomic energy and appropriate precautions were taken to protect the individual.

In particular, the members of the International Commission on Radiological Protection and the National Committee on Radiation Protection are to be commended for their foresight in developing standards and guidelines on radiation protection. More recently, the work of the Federal Radiation Council has been instrumental in stressing a need for a reasonable balance between benefits and risks. In its memorandum to the President, published May 18, 1960, on Radiation Protection Guidance for Federal Agencies, it states:

> "The fundamental problem in establishing radiation protection guides is to allow as much of the beneficial uses of ionizing radiation as possible while assuring that man is not exposed to undue hazard. To get a true insight into the scope of the problem and the impact of the decisions involved, a review of the benefits and the hazards is necessary."

Although the benefits from atomic energy are relatively easy to assess, the extent of hazard, if any, at low dose levels of chronic exposure is, in most cases, impossible to evaluate. The fact that there has been no evidence of injury from isotopes when used according to ICRP or AEC standards does not lessen our responsibility for controlling and avoiding unnecessary risks of radiation exposure.

This Note will discuss a program aimed at anticipating possible hazards from the many types of sealed sources of isotopes, as well as isotopic devices, and to perform tests aimed at providing information for more accurate assessment of the risks involved in permitting wide beneficial usage. Such information should lead to safer sources and devices and yet not cause unnecessarily strict standards for fabrication or use.

Safety testing program

Soon after passage of the 1954 Atomic Energy Act, 10 CFR Part 20, "Standards for Protection Against Radiation", based on NCRP recommendations, was developed and published as a proposed rule and has since become a regulatory standard. However, performance standards for sealed sources, under their various conditions of use, have never been established. This has been due largely to the lack of technical investigations aimed at this objective. Such information is essential to guide regulatory groups in establishing safe yet sensible regulatory practices, to assist the manufacturer in designing reliable equipment, and to assure the public that such equipment is built to meet proper safety-performance criteria. Test procedures and fabrication specifications presently used to evaluate the safety of sources and devices containing radioisotopes are those proposed by individual manufacturers. They are not standardized and do not necessarily represent conclusive studies on the parameters of safety under all conditions of use.

Recently, the AEC's Office of Isotopes Development, recognizing the increasing use of radioisotopes and the added possibility of release of radioactive materials to the environment through fires, accidents, etc., initiated a testing program to develop a technical basis for safety performance of isotope sources and devices. The interest of industry in this type of program was demonstrated during a recent meeting of sealed-source manufacturers in Washington. It was agreed by all those present that this program should be most useful to everyone concerned.

The program will include:

(1) Development and testing of radioisotope sources to assure:
   a. *Integrity of containment* of radioactive material within the sealed source capsule or imbedding matrix; and
   b. That, should the integrity of containment fail, the source material itself will constitute a *minimal risk of dispersal and/or biological absorption.*

(2) Development and testing of isotopic devices to assure:
   a. *Integrity of source within the device* against accidental loss or willful pilfering; and
   b. *Maintenance of an adequate degree of shielding* under probable conditions of use or accident.

To establish guidelines for such a program of safety testing, a survey study of all types of testing procedures has been initiated at Battelle Memorial Institute. We anticipate that this survey will result in an outline of research and testing efforts needed to fulfill the objectives of sealed-source and device testing. The National Bureau of Standards, the AEC's National Laboratories, and the Underwriters' Laboratories, Inc., all have special competency in the testing field and are expected to participate in the testing phases of this program.

Safety tests will be designed to determine the security of containment of sealed sources by subjecting them to a number of environmental, usage or accidental conditions, such as shock, mechanical stress, radiation damage, chemical effects, corrosion, fire, diffusion of radioactive material through the container or containment system, etc. Various types of capsule-closure techniques will be investigated, such as metal welding, ceramic coating, epoxy seals, material alloys and other measures to assure integrity of containment.

In addition to performance safety testing to assure *containment*, emphasis will also be placed upon "*inherent*" *safety in case of loss of containment*. This "second line of defense" can be achieved by the development of isotopic compounds or coating materials that are not easily dispersed or *are almost biologically inert*. It is believed this can be achieved without serious loss of source efficiency for performing intended purposes. Tests will be made to develop isotopic materials having maximum insolubility, heat resistance and other factors to minimize contamination or internal health hazard by any released radioactive material. In this regard, our Office sponsored the development and extensive testing of strontium-90 titanate for heat and radiation sources. This compound almost eliminates the biological uptake probability of this very useful, but otherwise "awesome" isotope.

Although the need for this type of program has long been recognized, its need is further emphasized by the growing interest in using radioisotopes in consumer items. These include luminescent devices, i.e. watches and instrument dials, aircraft markers for both civilian and military, exit markers in theaters and other public buildings, static control equipment, negative-ion generators, electron tubes and possibly many others.

Here we are faced with the fundamental problem of evaluating benefits versus risks, but in this case we have the condition of an extremely small radiation hazard of an individual item but multiplied by possible wide and unknown distribution. The particular isotope, how it is fixed chemically and physically in the item and the possible uses or misuses will help dictate the degree of release, if any, of radioactivity into the environment. To evaluate more fully the factors involved in weighing the over-all risks versus benefits, it is important to explore (1) the economics of the application, (2) the long time rate of usefulness, that is, will it continue to be useful at a given level or will the number of devices increase or decline because of popularity, and (3) the probability and rate of release of radioactivity to the biosphere and its contribution to the over-all (background plus man-made) exposure of the population.

The AEC recently exempted from licensing timepieces with up to 25 mc of tritium in luminous paint, provided the licensed manufacturer can apply the paint to timepieces in such a manner as to withstand specified tests to which the timepieces will be subjected. This action was unique in that it is one example of how atomic energy can reduce exposure to individuals by substituting tritium for radium which has been used for many years. Other devices using small amounts of radioisotopes intimately fixed in devices are under consideration for license exemption.

Studies are being conducted to develop criteria for assessing the possible contribution to the environmental background radiation as a result of large-volume distribution and eventual disposition of radioisotopes in such consumer items. The source safety testing program can help obtain some of the essential data regarding proposed items and investigate isotopic forms that may reduce possible risks.

Conclusion

The safety testing program for radioisotope sealed sources and devices is designed to develop a technical basis for more accurately assessing the health and safety aspects involved under conditions of use or accident. Such information will assist regulatory agencies in establishing safe yet reasonable regulatory standards and the manufacturer in designing more reliable equipment. Most important, the public will be assured that isotope equipment is built to meet carefully developed safety criteria.

Division of Isotopes Development
(formerly Office of Isotope Development) P. C. AEBERSOLD
U.S. Atomic Energy Commission J. W. HITCH
Washington 25, D.C.

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LETTERS TO THE EDITOR

Note on Suggested Regulations for Radiation Exposure

A RECENT issue of *Health Physics* (Vol. 5, Nos. 1/2, p. 99) contained a Note relating to the Suggested Radiation Protection Regulations developed by NCRP Subcommittee 10. The Note contained comments prepared by a Health Physics Society Committee on Radiation Regulations.* As Chairman of NCRP Subcommittee 10, I would like to reply to the comments made by this HPS Committee.

The HPS Committee states that it objects to the term "Health Physicist" in the revised regulations on the grounds that some radiation work is not complicated enough or hazardous enough to require the attention of a person having the competence now recognized as necessary in a Health Physicist. (In the original HB 61 and in other NCRP Handbooks the term "qualified expert" is used, and this term is also objectional to the HPS committee for the same reason.) Subcommittee 10 gave careful consideration to this matter in the preparation of the draft. It was agreed that it was not necessary to have a Health Physicist present on a full-time basis at every radiation location. Nevertheless, the Subcommittee felt that a Health Physicist should be consulted to establish the procedures and to check the equipment to be used in any radiation work. Certainly there are many cases where such expert consultation is required only when the project is started or at infrequent intervals thereafter. The Subcommittee carefully worded the appropriate paragraphs in the Suggested Regulations to permit this flexibility. It should be clear, however, that the Subcommittee felt it was necessary for all radiation work covered by the regulations to have *some* supervision by an expert.

The second suggestion made by the HPS Committee proposes that requirements for personnel monitoring be based on the probability of exposure. Specifically, it is suggested that routine personnel monitoring be required for persons who are likely to receive a dose in excess of some percentage of the maximum permissible exposure limits for external exposure. Such a rule would be completely unmanageable under most circumstances, since the permissible limit is based on the age and previous exposure of the individual. If personnel monitoring were required at some percentage of the permissible limit, it would be necessary to determine by the formula the exposure status of each individual each time he entered a controlled area. Having done this we would then be faced with a situation where personnel monitoring would be carried out for some persons but not for others who were doing identical work. The NCRP Subcommittee considered writing the requirements as suggested by the HPS Committee but finally decided it was impractical.

The third comment points out that the Suggested Regulations do not exempt personnel monitoring and record keeping for patients receiving radiation exposure for medical purposes. The Subcommittee intended to make this exemption and it certainly should be added to the regulations by anyone making use of them. Evidently the statement was inadvertently dropped out of one of the many drafts and none of the Subcommittee noticed the deletion.

The fourth comment deals with radioactive waste disposal and involves many problems which cannot be discussed satisfactorily in a short note such as this. The HPS Committee feels that the limits in Section 13 (b) (2) may be unnecessarily restrictive for some situations. The Subcommittee believes that where this is so the user should be able to persuade the Agency of the fact and obtain relief. In this section of the regulations, the Subcommittee tried to provide adequate waste control for all situations and a means for obtaining relief from the numbers where these could be demonstrated to be too restrictive. As far as the special exemption on excreta of patients is concerned, I can see no reason at all to consider this radioactive waste any different from radioactive waste coming from other sources, and therefore find it hard to justify less control of such wastes as the HPS Committee recommends.

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(Chairman, NCRP
Subcommittee 10)
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Schenectady, N.Y.

* *Editor's comment:* It has been determined that this note was an expression of opinion of only this Committee and did not necessarily represent the views of the Society Membership or its Officers.

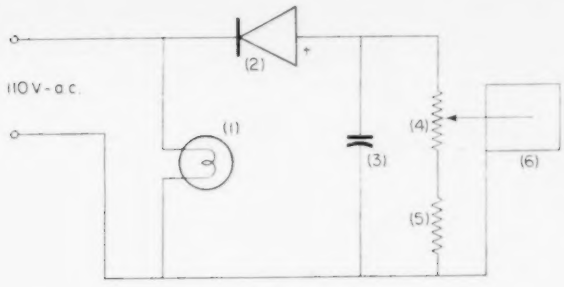

FIG. 1.

Conversion of a Landsverk Model L-21, Pocket Dosimeter Charger for Use on a 110-V a.c. Circuit

(*Received* 27 *June* 1961)

THE replacement of batteries in pocket dosimeter chargers after each six months of operation (battery cost = $8.00) has resulted in the design of a circuit to modify the Landsverk, Model L-21, charger for operation on a 110-V a.c. circuit. The circuit, designed specifically for the Landsverk L-21, could be used for other types of chargers. The cost of materials for the modification is approximately $3.00. The major components, as shown in Fig. 1, are: (1) 6S6/6W6 pilot lamp placed under dosimeter well so that scale is visible; (2) 110-V a.c. (min) silicon rectifier; (3) 1-μF 200-V d.c. (low leakage) capacitor; (4) 10-MΩ, ½-W potentiometer; (5) 10-MΩ, ½-W resistor; (6) well for inserting pocket dosimeter.

Engineering Experiment Station D. K. DAVIS
Georgia Institute of Technology

Considerations of Radioactive Fallout in Livorno (Italy)

(*Received* 18 *July* 1961)

RADIOACTIVE fallout is collected on a monthly basis at Livorno. The period of collection is one month or one fortnight and is made by means of a high-walled polyvinyl chloride pluviometer, periodically washed in distilled water.

The bottom of the container is connected to a cartridge made of a column of cellulose pulp and two columns of ionic exchange resins, first cationic then anionic, through which the water of the container flows with an average flux of 1 l./hr. At the end of the sampling the cellulose pulp and the resins are reduced to ash at a temperature of approximately 350°C. Then the whole beta activity of the residue is measured by means of a scintillation counter furnished with plastic phosphors. Finally, the samples of the preceding month are measured.

In Fig. 1, the amount (in mc/km^2) of radioactive fallout deposited on the ground at the Livorno station is plotted as a function of time. Curve no. 1 represents the radioactive decay of the whole deposit relating to collections made during the months from July to December 1959.

The broken line on 31 January 1960 corresponds to the increment of active fallout in January, while curve no. 2 is obtained by adding to curve no. 2, the experimental values of radioactive decay of the sample collected in the same month of January.

Proceeding in an analogous way, the curve family has been obtained.

Many radioactive products are present. The mass of measurements shows the prevalence of two groups: one of short half-life of the order of 60 days, and one of long half-life of the order of 2 years, so that the curves are fairly well represented by the equation:

$$A = ae^{-1.31\times10^{-3}t} + be^{-1.46\times10^{-3}t}$$

It is deemed more convenient to represent the same decay as was done in the generical case of fallout with the following function:

$$A = 76.6t^{-0.37}$$

where t is measured in days from the first day of each curve and A is the radioactivity on the t day.

Curves 3, 4 and 5 refer to samples of fallout collected during the first fortnight of the respective month. They are interpreted in correspondence to the day in which the same samples of fallout were added to those picked up during the second halves of the respective months. Decay of the sum is represented respectively by curves 3, 4 and 5.

The greatest increment to fallout accumulated on

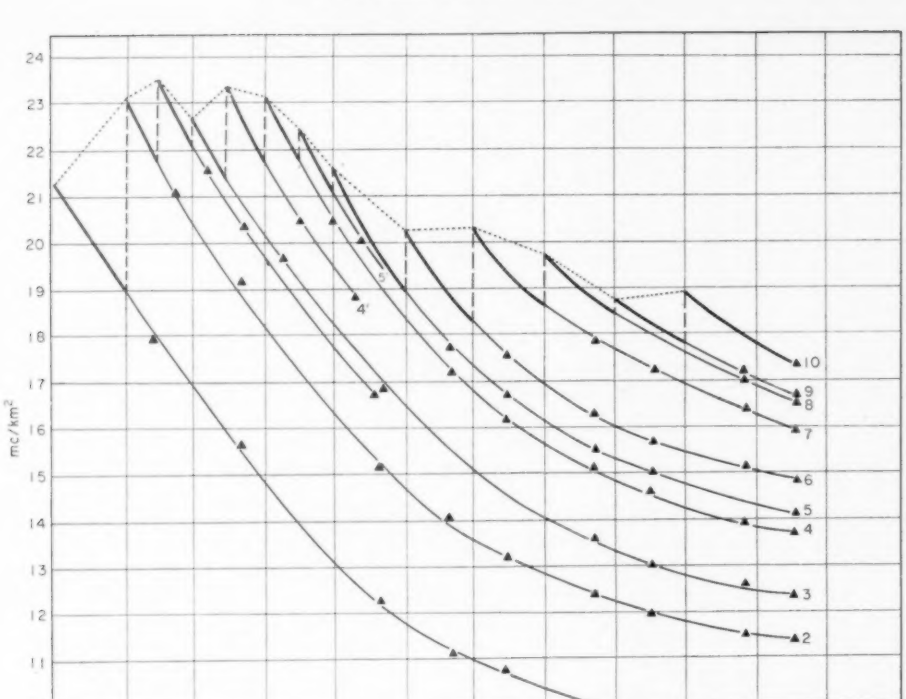

FIG. 1. Radioactivity vs. time.

the ground level occurred in January 1960 and had a value of 4.25 mc/km². Additions of 2.37 mc/km² and 3 mc/km² were made respectively in February and March 1960 while the fallout deposited during the remaining months was of about 1 mc/km² per month.

The greatest washing of the atmosphere by rain (108 mm in 11 days) occurred during January 1960. It must be noted that in April, in spite of a heavier rainfall, the quantity of radioactive fallout was smaller than that of February. This can be explained by considering the fact that not only the whole amount of rainfall during the month is important but also the number of rainy days, because radioactivity is in the greatest part carried in the first rain fraction.

Table 1 shows that radioactivity is a function of both the number of rainy days and the depth of rainfall.

Actually radioactivity is deeply influenced by how the rain has fallen, namely, the amount fallen in each period, and the interval among the various rainy periods. In fact, during May with only one day of rain and a rainfall of 0.5 mm, there was, in a relative sense, the greatest increment in radioactivity. The radioactive fallout found undoubtedly comes from the stratospherical reserve created by the explosions which had taken place until October 1958.

Table 1

Sampling period	Curve	Radioactivity (mc/km²)	Rainfall (mm)	Rainy days
January	2	4.25	108	11
February	3	2.37	54.5	11
March	4	3	88	12
April	5	1	71	7
May	6	1.25	0.5	1
June	7	2.1	17	2
July	8	0.90	12.5	4
August	9	0.20	0.5	1
September	10	1.20	47.7	5

The low radioactive amount is in accordance with a medium period of stay in the stratosphere of approximately 1 year.

The dotted curve should represent the actual development of the fallout if collection of fallout were made daily. It would follow that, with a constant daily effort, we can write, in a purely theoretical way, that

$$A = Bt^{0.63-K}$$

which, for January becomes

$$A = 113.5t^{-0.41}$$

where t is measured in days from the beginning of each collection. As a matter of fact, after an increase in radioactive fallout until March 1959, it has been decreasing because the monthly increment has been constantly lower than the natural decay.

Acknowledgment—We wish to thank Prof. T. FRANZINI for his valuable advice and suggestions.

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THE EFFECT OF PHARMACOLOGICAL DOSES OF NON-RADIOACTIVE IODIDE ON THE COURSE OF RADIOIODINE UPTAKE BY THE THYROID

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(*Received* 16 *March* 1961; *in revised form* 12 *May* 1961)

Abstract—Doses of potassium iodide, containing 200 mg of iodide, inhibited further thyroid uptake of radioiodine in six subjects when given 2½ or 4 hr after single oral doses of radioiodine.

It is known that radioactive iodine, if ingested in the form of iodide, is taken up by the normal thyroid during the ensuing hours, the maximum uptake being reached after 1 or 2 days and half this maximum value after approximately 6 hr. It has long been known that the extent of this uptake is reduced if stable iodides are given at the same time as the radioiodine.[1] It seemed useful to find out whether the progressive uptake could be slowed or halted, and if so, how rapidly, if a large dose of stable iodide was given a few hours after an oral dose of radioiodine. Accidental contaminations with radioiodine could be of importance following ingestion or skin puncture in laboratories dealing with therapeutic quantities of this isotope, or from ingestion or inhalation by members of the population following accidental emission of fission products from a nuclear reactor under conditions such as occurred at Windscale[2] in which radioiodine was the radioelement of greatest importance.

The action of a pharmacological dose of iodides in arresting the uptake of radioiodine by the thyroid was examined in six normal subjects after doses of 15–25 μc of I^{132} given orally in the form of iodide. The thyroid radioactivity was determined by profile counting using a plastic scintillation counter. Counts were made at times (Table 1) before and after a dose of potassium iodide given in water and containing 200 mg of iodine, or of sodium chloride containing 200 mg of chlorine in control runs made on separate days.

This dose of iodide was selected for trial since it is of conventional size in normal medical use, comparable or larger doses commonly being given several times daily for long periods in expectorant cough medicines, and rarely causing toxic effects. At the same time it contains about 1000 times the normal daily intake of iodine and so might be expected to inhibit further uptake fully and perhaps rapidly.

In estimating the thyroid uptake, allowance was made for the radioiodine content of neck tissues by a correction based on the counting rates observed at points on the profile curve adjacent to the peak opposite the thyroid. Mean values (Table 2) were observed for the thyroid content, expressed as percentages of those at the time of the doses of NaCl or KI, and corrected for decay.

Renal clearances were not determined, but it was clear that the excretion of radioiodine was not arrested by the dose of iodides, the excretion rates during the 3¼-hr period following the doses of iodides having averaged about 65 per cent of those observed during the initial 2½ or 4 hr following the radioiodine and before iodides were given. In the control subjects, the thyroid uptake rate during the period of 3 hr after the administration of chlorides averaged 45 per cent of that during the 2½ hr period after the radioiodine dose.

Table 1

Subject	I^{132} (μc at 0 hr)	Profiles at (hr)	Dose at (hr)	Profiles at (hr)	Urine at (hr)
Controls					
D, E, G	15–19	1.6, 2.4	NaCl at 2.5	3.6, 4.6, 5.6	
Given iodides					
D, E, W	25–27	1.6, 2.4	KI at 2.5	3.6, 4.6, 5.6	2.5, 5.8
G, S, X	22–25	1.8, 2.8 3.8	KI at 4.0	5.1, 6.1, 7.1	4.0, 7.2

The arrest of further uptake by 200 mg of iodide was confirmed in one further subject with chloride or iodide given at 3 hr after I^{132}, and with profiles continued for a further 5 hr; and in two patients given 250 mg of iodide at 3 and at 4 hr after I^{131} (without control runs) and followed by profile counts for a further 2 hr, with a further profile 24 hr later which showed a thyroid content no greater than at the time of the iodide doses. The correction for the radioiodine content of neck tissues, and the decrease of this content with time, were examined with the same profile counter technique in three athyreotic subjects, who had no radioiodine uptake in the thyroid and no detectable tumour tissue in the neck after radioiodine treatment for thyroid carcinoma.

A small discharge of the radioiodine already concentrated in the gland at the time of the iodide dose cannot be excluded. The low uptake of the thyroid within $2\frac{1}{2}$–4 hr of the test dose makes it difficult to allow for the content of neck tissues with precision, and the profiles after two or three half-periods of I^{132} decay were not obtained with high accuracy. It is possible, therefore, that a progressive fall in total neck radioactivity which was observed after KI (after correction for decay) is due to the fall in the radioiodine content of neck tissues other than the thyroid, with a constant thyroid content, rather than to any fall in the content of the gland itself.

Table 2

Subjects	At time of dose of:	At 1.1 (hr later)	2.1 (hr later)	3.1 (hr later)
Controls				
D, E & G	NaCl 100	128	149	154
Given iodides				
D, E, W	KI 100	97	87	100
G, S, X	KI 100	99	94	96

A simple arrest by iodides of further uptake of radioiodine would, however, have the effect of reducing the radiation delivered to the thyroid by a factor of between 5 and 3, if iodides were given at an interval of 2–3 hr after absorption of radioiodine. It would therefore appear that, after an accidental contamination involving substantial amounts of long-lived radioiodine, the administration of stable iodides would still be of value even if only given a few hours subsequently.

Acknowledgments—We have appreciated the opportunity of discussing the importance of methods for arresting radioiodine uptake by the thyroid with Professor J. R. Squire, and with Mr. C. A. Adams and Dr. J. A. Bonnell.

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ADMINISTRATION OF STABLE IODIDE AS A MEANS OF REDUCING THYROID IRRADIATION RESULTING FROM INHALATION OF RADIOACTIVE IODINE

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(*Received* 27 *February* 1961; *in revised form* 20 *May* 1961)

Abstract—Despite the emphasis placed on safety considerations during the design and operation of nuclear power stations, it is prudent to consider the possibility of a reactor accident capable of causing radiological injury to the general public residing in the vicinity. This implies the emission of radioactive material from the reactor. The first danger which must be reduced in connexion with certain types of reactor is that resulting from inhalation of radioactive materials by persons in the path of the cloud. All existing evidence suggests that for the type of reactor under consideration, the inhalation of radioactive iodine is the most immediate hazard. Iodine is selectively absorbed and concentrated in the thyroid gland where it is retained for relatively long periods. In this report an analysis is given of the factors governing the uptake of iodine by the thyroid gland, and, by using data available in the literature, it is shown to be probable that the ingestion of small amounts of inorganic iodide would significantly reduce the dose due to concurrently inhaled radioactive iodine.

It is therefore advocated that the administration of stable iodide should be seriously considered as an alternative or supplement to evacuation of any persons at risk from inhalation, following a reactor accident. The adoption of this suggestion would simplify emergency plans by reducing the need for evacuation or at least add valuable time during which evacuation could be better organised.

I. INTRODUCTION

THIS paper is prompted by consideration of the circumstances which would arise in relation to power reactors now under construction in the United Kingdom if there were reason to suspect that an accidental release of fission products had occurred from them on a sufficient scale to cause an inhalation risk to individuals downwind. It is well known that the accident at Windscale No. 1 Pile in 1957 resulted in a release of radioactive iodine in a far greater proportion than that corresponding to its concentration in normal fission products.[1]

FLETCHER and FARMER[2] using results given by DUNSTER *et al.*[3] emphasize that the preferential releases of iodine and caesium with respect to strontium are consistent with the relative volatilities of the elements. The results of a further assessment of the release are given in an appendix to a report[4] to the Medical Research Council by a Committee under the chairmanship of Sir HAROLD HIMSWORTH. These results with the melting and boiling points of the elements concerned, are shown in Table 1.

It should be stressed that throughout this paper consideration is confined to those releases of radioactive material which could conceivably occur from gas-cooled graphite-moderated

Table 1

Element	Estimated release (c)	m.p. (°C)	b.p. (°C)
Iodine-131	20,000	114	184
Tellurium-132	12,000	452	1007
Caesium-137	600	30	690
Strontium-89	80		
Strontium-90	2	770	1300

natural uranium reactors. The power reactors of this type now under construction by the Central Electricity Generating Board differ from the Windscale piles by using a self-contained gas-coolant circuit, with carbon dioxide instead of air as the coolant. An accident similar to that at Windscale therefore could not occur with these civil reactors. The possibilities of other types of accident have been studied. All forseeable contingencies which could result in dangerous releases of radioactivity from the reactors are covered by safety precautions in the design and operation of the stations. These measures are nevertheless not assumed to remove the responsibility to prepare emergency schemes which would minimize the radiological exposure resulting from an accident.

In their report, FLETCHER and FARMER[2] imply that the first action which would be required in an emergency is the evacuation of persons occupying that region in a downwind direction within which it would be possible for inhalation of radioactive iodine to lead to a thyroid dose exceeding 25 rems, the relevant limit recommended by the Medical Research Council[5]. The policy of siting nuclear power stations in regions remote from large aggregations of population ensures that any evacuation which might be necessary would involve only a small number of people and would therefore be practicable. However, evacuation, even though it may last only for several hours, would necessarily cause discomfort and anxiety and could lead to alarm and to secondary accidents. It would therefore be a disservice to put such emergency plans into action in circumstances which were later shown to be not sufficiently serious to justify evacuation.

In any accident leading to a release of fission products, those in control of a nuclear power station will require reliable information on downwind concentration of I^{131} and good forecasts of the likely total release and its distribution for some hours ahead. An accurate assessment will take time. On the other hand, if the initial assessment of the situation indicates that there may be danger in delaying evacuation there would be an obligation to put it in hand as soon as possible. The situation would be eased if a method were available of increasing the time for assessment without thereby increasing the risk of causing an excessive thyroid dose.

An explosive release of a large quantity of fission products can be regarded as incredible. Any credible but still highly unlikely accident would therefore afford time in which possibly some safeguarding action other than evacuation could be taken. The thesis of this paper is that the administration of stable iodine, to those individuals whom it would be necessary to evacuate if the pessimistic forecast of conditions proved correct might provide a simple and safe means of reducing irradiation of the thyroid. If this action were taken as an "alerting" action, the need for evacuation might be reduced or eliminated.

This conclusion follows primarily from the fact that more prolonged inhalation could be allowed before the resulting thyroid uptake would reach any permissible limit which might be adopted. Added advantages are that action would not have to be based only on a quick assessment. Where rapid decisions are necessary it would be natural, and justifiable, to overestimate the "danger area" and the duration of the release.

It is well established that large quantities of inorganic iodine ingested before the administration of radioactive iodine can very substantially reduce the thyroidal uptake of the latter. CHOU *et al.*[6] found that the uptake of I^{131} by the thyroid was less than 1 per cent of the administered dose in patients who had received ten drops of Lugols iodine three times daily for 24 hr (i.e. an aggregate of 260 mg of iodine) before receiving a dose of 300 μc of I^{131}. If administration of some form of stable iodine is to be used in emergency conditions it is necessary to have numerical estimates of the relation between the quantity of stable iodine, the time at which it is administered and the dose reduction. These aspects are considered in detail in later sections of this paper. A simplified treatment which indicates some quantitative aspects is given below.

II. DEPENDENCE OF THYROID RADIATION DOSE ON THE AMOUNT OF AVAILABLE IODINE

The radiation dose to the thyroid in any individual case depends on the ability of the

gland to take up and retain radioactive iodine from the circulating blood. If the thyroid has a limited capacity for iodine retention, the uptake of radioactive iodine will depend upon the total iodine available to it in the bloodstream. A crude estimate of the "uptake" of radioactive iodine can be made if the following circumstances are assumed:

(1) The bloodstream contains a mass M of available iodine, M being large compared with the normal iodine content of the bloodstream.

(2) The thyroid is capable of absorbing iodine additional to its "normal" content only up to some limit W.

If radioactive iodine is administered concurrently with the mass M of stable iodine, the fractional uptake of radioactive iodine (assuming the secretion during uptake to be negligible compared with W) will at most be W/M. The consequent radiation dose to the thyroid will thus be approximately proportional to this fraction, or, if percentages are used, to 100 W/M.

The uptake as normally defined and measured is a percentage activity A which commonly lies between 20 and 40 per cent. If a value of about 100 mg is assumed for M, the ratio of the radiation dose to the thyroid in the circumstances governed by (1) and (2) to the "normal" dose will thus be less than W/A.

The value of W which applies in any individual case will probably vary with A. A reduction in dose will be achieved by administering 100 mg of stable iodine if W is less than A. Thus, if A is arbitrarily taken as 30 per cent for an average case, a gain is certainly introduced if W is less than 30 mg. (This statement does not imply that there is no gain if W is less than 30 mg.) It seemed to the authors to be probable that the amount of iodine which could be stored by the thyroid (above its normal complement) would, in normal cases, be considerably less than 30 mg, probably by a factor of at least 10. Since a search of the literature did not provide numerical values for W, the authors, by the courtesy of Dr. STUART MASON, obtained an indication of the values in their own cases. The data are given in Appendix I. The salient points are summarized in Table 2.

Table 2

	A (% uptake in absence of carrier)	A' (% uptake with 100 mg KI)	A'/A
C.A.A.	30	3	0.1
J.A.B.	38	4	0.1

Since the subjects of the experiment represent a random choice so far as the thyroidal properties are concerned, the results illustrate a probability that W is small compared with 30 mg. These examples are no more than illustrative and the tests were in fact made some time after the theoretical investigation given in Appendix II had been made.

Several approximations are involved in assuming the dose to the thyroid to be proportional to the "uptake". For example, the dose during uptake and the effects due to recycling of iodine are neglected. These effects seemed to be likely to have only a small effect on the total dose. In order to estimate whether the approximation was justifiable a more detailed theoretical analysis was made. This is described in later sections of this paper and in Appendix II.

III. THE THREE COMPARTMENT MODEL OF THE IODINE REGIME

A more realistic analysis of the dependence of thyroid dose on the amount of available iodine can be made by using a model based on that described by STANBURY *et al.*[7] The use of a model, analysed in mathematical terms, may at first sight appear to import an unjustifiable degree of precision into the subject. However, numerical predictions of the doses which are likely to be given must inevitably be made even if the only question involved is that of evacuation. Some form of theory relating air concentrations to the consequent thyroid dose is necessarily implied when a decision is made on this issue at the time of a release. Once the fairly obvious idea is brought into consideration that "blocking" by stable iodide may be an effective method of reducing thyroid dose the equally obvious question arises "how effective"? A simple

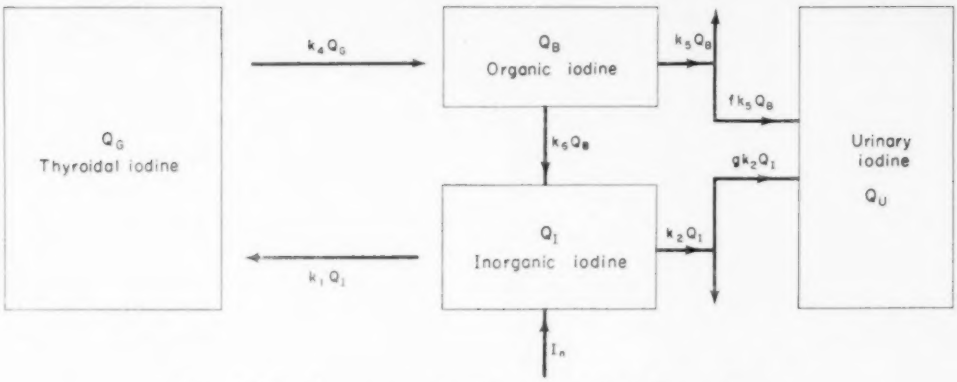

FIG. 1. The three-compartment model of the iodine regime.

qualitative consideration of this question does not provide a useful answer and in any case is open to the charge that the problem is over simplified. A more detailed analysis of the relevant studies which have been made is in the authors' view most readily undertaken by using mathematics as a tool to find the consequences of certain simple assumptions. The non-mathematical reader may find the analysis elaborate, but it is suggested that the technique of reaching the conclusions adds no complication to the simplicity of the postulates. Measurements are available in this field and it is commonplace to represent them in the form of graphs. The extension to representing them in the form of equations is again no novel feature. The mathematics can be regarded as a compact method of expressing a description of certain measurable features. There is no suggestion in using the model that new facts about the chemistry of iodine uptake or transfer are revealed. Similarly there is no suggestion that individual studies will exactly confirm the equations which are developed. Scatter in any individual results is to be expected. The model is regarded as having the same sort of usefulness as many of the calculations made in the domain of Health Physics and concerned with the approximate estimates of doses.

The model used is illustrated in Fig. 1. Its first characteristic is that it assumes that any iodine taking part in the regime can be regarded at any given time as either:

(i) located in the thyroid and available for conversion to and transmission as organic iodine

or (ii) in inorganic form outside the thyroid and available for transfer to the thyroid or for excretion

or (iii) in organic form outside the thyroid and available either for conversion to inorganic form or for excretion.

Its second characteristic is the assumption that the rates of transfer from compartments are proportional to the amounts of iodine in the compartments. The rate constants (k_1, k_2, etc.) together with the iodine contents (Q_I, Q_G, Q_B) thus determine the transfer rates at any specified time. When analysis of the behaviour of the model is applied to estimating the behaviour of radioactive iodine it is necessary to make a third assumption. This is that the concentration of radioactive iodine throughout any one compartment is uniform at any instant and adjusts itself without any significant time delay to the concentration required to accommodate the addition of iodine having a different proportion of radioactive iodine.

These simple assumptions may be far removed from the complex situation which actually exists. However the broad consequences, as indicated by measurements of thyroid and urine activity, appear to be consistent with formulae deduced from analysis of the model, e.g. by STANBURY *et al.*[7] and by RIGGS.[8] The analyses of which the authors are aware have introduced approximations into the mathematical solutions. These approximations are justifiable for some ranges

Table 3

Symbol	Definitions
I	iodide compartment
G	thyroid organic iodine compartment
B	extrathyroid organic iodine compartment
Q_I, Q_G, Q_B	quantity of iodine in I, G, B at time t
Q_I', Q_G', Q_B'	quantity of radioiodine in I, G, B at time t
Q_I^x, Q_G^x, Q_B^x	quantity of radioiodine which would be in I, G, B at time t in the absence of radioactive decay
Q_u, Q_u', Q_u^x	quantity of inactive, radioactive and labelled iodine ("non decaying") accumulated in the urine at time t
I_n	rate of intake of inorganic iodine to the I compartment from external sources
k_1	rate constant of accumulation of iodine by G from I
k_2	rate constant of total excretion of iodine from I
k_3	rate constant of radioactive decay of radioiodine
k_4	rate constant of loss of iodine from G
k_5	rate constant of total excretion of iodine from B
k_6	rate constant of delivery of iodine from B to I
f	fraction of excreted organic iodine which goes to urine
g	fraction of excreted inorganic iodine which goes to urine
P	fraction of total excreted iodine which goes to urine
β	fraction of urinary iodine in organic form
t	time (unless otherwise stated t is measured from the time of administration of radioiodine)

of numerical values, including those most commonly encountered. However the full solutions can readily be obtained by standard mathematical methods, and thus give formulae which are valid for any range of the constants. These are given in Appendix II. The symbols used in the analysis are defined in Table 3.

The quantities defined in Table 3 are not all independent. One relation follows as a matter of definition, viz. $Q' = e^{-k_3 t} Q^x$ when t is measured from the time at which $Q' = Q^x$. Q' is required when doses are calculated and Q^x when theoretical and experimental results are compared.

The above symbols are sufficient for a description of the three-compartment model in which the k's remain constant. In a later part of the paper the hypothesis is put forward that k_1 is not a constant. The symbols suitable to a treatment of this case are defined in Section V.

IV. CONCLUSIONS FROM THE MODEL IF THE RATE CONSTANTS DO NOT VARY

If the rate constants for any individual are constant over the period of time concerned, the dose to the thyroid resulting from the administration of a quantity $(Q_I^x)_0$ to the bloodstream is proportional to a quantity I, the expression for which is derived in Appendix II [equation 31].

The "uptake" U can be defined as the maximum thyroid content of radioactive iodine (without allowance for radioactive decay). It can readily be shown that, taking into account the likely relative magnitudes of the constants, the model leads to a value of U given by

$$U \simeq \frac{k_1 (Q_I^x)_0}{k_1 + k_2}$$

I can be expressed approximately as

$$I \simeq \frac{U}{k_3 + k_4}$$

This is the value which would be obtained directly from the simple approximation that the thyroid takes up the quantity U in negligible time (from a total dose aspect) and loses it by radioactive decay and secretion at the rate characterized by k_4.

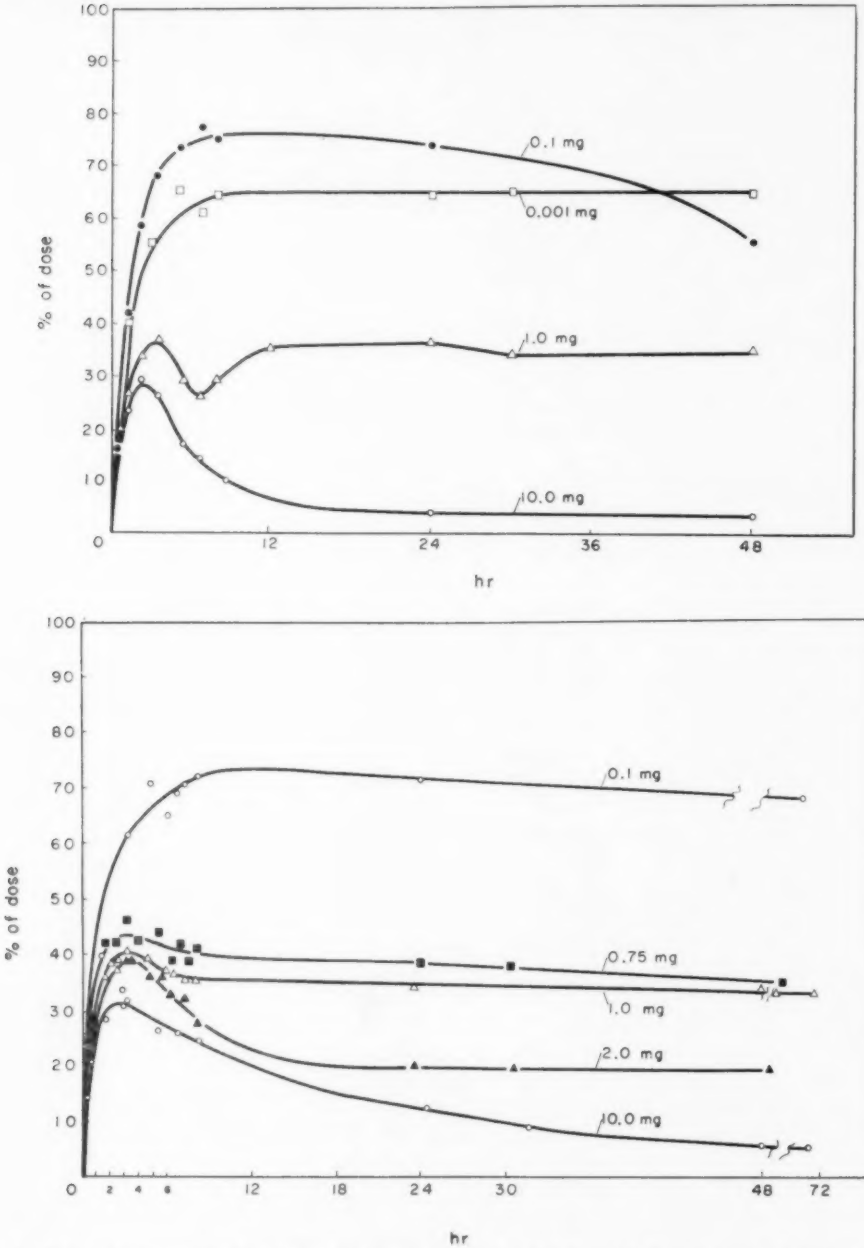

FIG. 2. *In vivo* observations of radioactive iodine uptake in the thyroid in two cases of exophthalmic goitre. (After CHILDS *et al.*[9]) (a) Thyroid I^{131} uptake curves following doses of 0.001, 0.1, 1.0 and 10.0 mg of stable iodide. The urinary excretion curves obtained in this patient are reproduced in Fig. 3.
(b) In this instance 0.1, 0.75, 1.0, 2.0 and 10.0 mg of stable iodide were given with the radioactive iodine tracer.

The model leads to formulae for the radioactive iodine content of the various compartments and of the urine. These are shown in Appendix II to be consistent with the simple approximations which can be applied at times in the region of several hours after the radioactive iodine has been administered. The approximations for the thyroid and urine contents are

$$Q_G{}^x \simeq \frac{k_1 (Q_I{}^x)_0}{k_1 + k_2} (1 - e^{-(k_1+k_2)t})$$

$$Q_u{}^x \simeq \frac{g\, k_2 (Q_I{}^x)_0}{k_1 + k_2} (1 - e^{-(k_1+k_2)t})$$

All the above formulae, and also the "exact" solutions of Appendix II demonstrate a feature of the model which is important in the present connexion. This is that the radioactive iodine contents of the various compartments (and hence the doses to any of the organs) are, on the postulates of the model, independent of the amount of stable iodine. The reason lies in the assumption that the rate constants (i.e. the k's) are independent of the amounts of iodine in the various compartments (i.e. the Q's). Constancy of the k's implies that an increased component of stable iodine will increase the transfer kQ from one compartment to the next because Q is increased. However the concentration of radioactive iodine in the total iodine transferred will diminish in the same proportion as Q is increased and hence the quantity of radioactive iodine transferred will remain unaltered.

For circumstances in which there is no abnormal variation in the total iodine available the assumption would not be expected to lead to any inconsistencies with experimental results. If small variations occur in the k's it will still be possible to regard a mean value as applying over the period of observation. This conclusion is consistent with the following statement by STANBURY *et al.*[7] "The effects of graded quantities of carrier iodide on thyroid function in thyrotoxic man have been studied by CHILDS *et al.* They found that there was no change in the proportion of iodide which was accumulated by the gland when the quantity of carrier iodide varied from a negligible amount to 100 μg." Subsequent parts of the same reference and observations reported by CHILDS *et al.*[9] and KEATING and ALBERT[10] show that this independence no longer obtains when large deviations are made from equilibrium conditions. The primary purpose of this paper is to provide an estimate of the alterations in the thyroid dose when a large quantity of stable iodine is administered concurrently with the inhalation of radioactive iodine. In the following sections the suggestion is made that this estimate can be made by using a simple formula to express the variation of k_1 (the uptake rate coefficient) with Q_G (the amount of iodine in the thyroid). The three compartment model is retained and the results are shown to be in broad agreement with those observations known to the authors which relate to cases of large iodine excess or deficiency.

V. THE DEPENDENCE OF THE THYROID UPTAKE RATE ON THE THYROID IODINE CONTENT

An enhanced uptake rate in cases of chronic iodine deficiency is evident from a large number of cases quoted by STANBURY *et al.*[7] and from the numerical values given by RIGGS.[8] A corresponding reduction in thyroid "uptake" when large quantities of carrier iodine are used is shown by the curves of Fig. 2, reproduced from the paper by CHILDS *et al.*[9] An increase in urine activity as the carrier iodine increases is shown in Figs. 3 and 4 reproduced respectively from the papers by CHILDS *et al.* and by KEATING and ALBERT. The evidence from the thyroid curves of Fig. 2 is direct in showing a reduced uptake. It can be seen immediately that this effect could be explained, in qualitative terms, by the assumption that k_1 diminishes as Q_G increases. The introduction of a large amount of iodine into the I compartment will tend to produce an increasing amount of iodine in the thyroid. If the latter increase causes k_1 to diminish, the mean value of the rate constant k_1 will be less than the value k_1 which corresponds with the approximate equilibrium existing before the large amount of stable iodine was administered.

Although the evidence provided by the urine curves is less direct, it can readily be shown that the results are consistent with the same hypothesis. First it is to be noted that both the urine

and the thyroid curves are insensitive to small additions of carrier iodine, up to about 0.1 mg. Since the thyroid may be assumed to contain several milligrams of iodine, the percentage increase in its content resulting from this increment in the I compartment would be expected to be small. Hence any consequent effect on the thyroid uptake rate would be expected to be correspondingly small. There will therefore be no significant difference made to the radioactive iodine content of the I compartment and hence if k_2 remains constant the urine activity will also be virtually unaffected.

The urine activity during a period of some hours after administration of the iodine can be approximately represented by

$$Q_u^x = \frac{g\,k_2\,(Q_I^x)_0}{k_1 + k_2}\,(1 - e^{-(k_1+k_2)t})$$

The apparent maximum to which Q_u^x tends is thus $(g\,k_2/k_1 + k_2)\,(Q_I^x)_0$. This apparent limiting value thus increases as k_1 decreases. The assumption that a large quantity of carrier iodine will lead to a small (mean) value of k_1 is thus consistent with the curves of Figs. 3 and 4. These show that up to about 48 hr the 10-mg curves appear to be approaching a very much higher limiting value than the 0.1-mg curves. At any given time the above equation implies that the slope of the urine curve will increase as k_1 diminishes. In terms of the hypothesis this conclusion corresponds with an increase of slope as the carrier increases. This feature is also observable in the curves.

The hypothesis is obviously consistent in general terms with the observed effect that a high uptake rate occurs in cases of iodine deficiency. Hence there appears to be good justification for a more detailed examination of the consequences of the assumption that, in the three-compartment model, the "constant" k_1 diminishes as the thyroid iodine content increases.

To avoid confusion, a new symbol λ will be used to replace k_1, defined as follows:
Rate of transfer of iodine from the I to the G compartment $= \lambda Q_I$. The simplest assumption to make, consistent with the type of

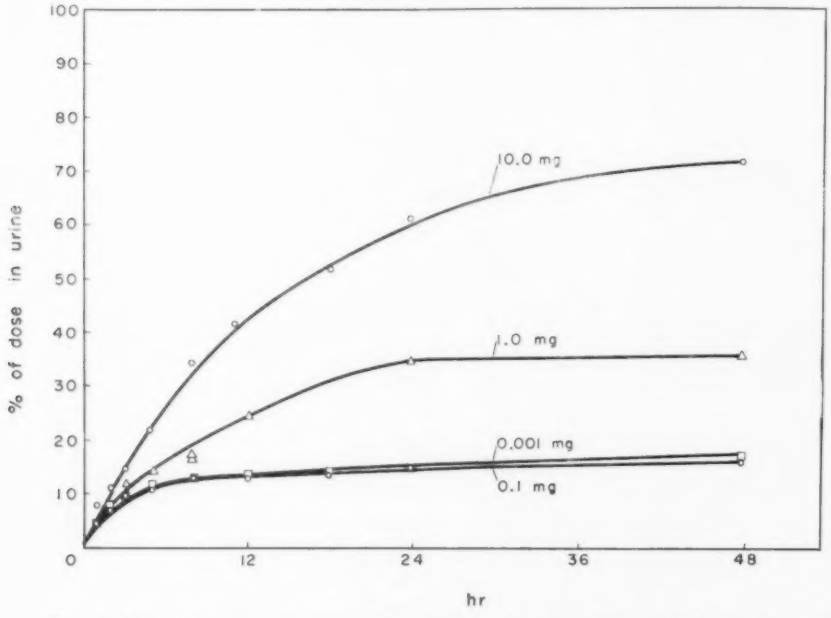

FIG. 3. Urinary excretion curves of radioactive iodine, the same patient as Fig. 2 (a).

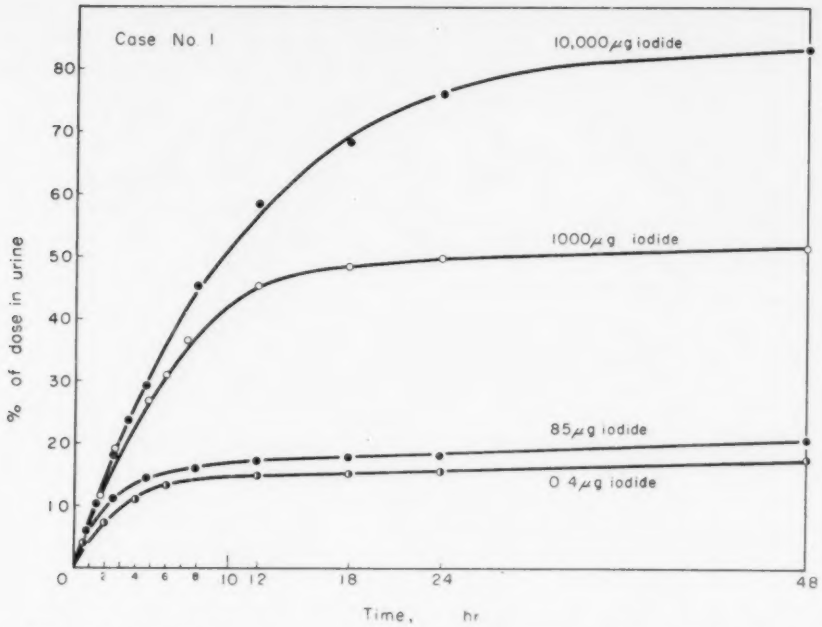

FIG. 4. Effect of variation of carrier iodide on urinary excretion of radioactive iodine. (After KEATING and ALBERT.[10]) With the larger quantities of iodide carrier, larger increments of radioactive iodine appear in the urine and the excretion of significant amounts persists for longer periods. Quantities greater than 10,000 γ (i.e. 10 mg) did not appear to increase the percentage urinary excretion much further.

variation discussed above, is that λ is a linear function of Q_G, i.e.

$$\lambda = R(1 - Q_G/Q_S)$$

where R = notional rate constant which would apply if the thyroid were completely devoid of iodine ($Q_G = 0$) and Q_S = notional mass of iodine which, if it could be present in the thyroid would reduce the uptake rate to zero ($\lambda = 0$).

Let λ_m = rate constant corresponding with an equilibrium in which the mass of iodine in the I compartment is maintained at mean value m,

and λ_M = rate constant corresponding with an equilibrium in which the mass of iodine in the I compartment is maintained at mean value M.

It is shown in Appendix II that if, at $t = 0$, the mass of iodine in the I compartment is changed from m to M, the above assumptions imply that λ departs from the value λ_m and asymptotically approaches the value λ_M, the time constant of the change being $\lambda_M/R\,k_4$. Using the above results it is possible to obtain an approximate solution for the activity in the urine as a function of time, for the periods covered by the observations illustrated in Figs. 3 and 4. As shown in Appendix II this analysis shows that the deductions from the model agree with the main features of the experimental curves.

VI. TOTAL DOSE TO THE THYROID WHEN THE UPTAKE RATE VARIES

A simplification is made in the analysis given in Appendix II of the radiation dose to the thyroid resulting from a variable uptake rate. Expressed in physical terms the simplification may loosely be described as neglecting the recycling of radioactive iodine which becomes available by extrathyroidal conversion of organic to inorganic iodine. A solution of the full equations is shown in Appendix II to be unnecessary in defining, in terms of the model, an

upper limit to the dose rather than the "exact" value. Hence, in the following, the doses discussed are those which would be given if there were no conversion from organic to inorganic iodine.

In the application to emergency schemes it must be assumed that some radioactive iodine has been inhaled before any remedial action, whether by evacuation or by administration of stable iodine, can be effected. Let

$(Q_G')_0$ = amount of active iodine already in the thyroid at $t = 0$,

$(Q_I')_0$ = amount of active iodine additionally introduced into the bloodstream at $t = 0$

(In fact there will be a continuing accumulation in the bloodstream for as long as the inhaled air contains active iodine. By assuming that all the activity is given at one instant the dose is slightly overestimated, and hence the simplification errs on the 'safe' side.) Let

D_1 = dose resulting from initial activity $(Q_G')_0$

D_2 = dose which would be given by $(Q_I')_0$ if $\lambda = \text{constant} = \lambda_M$

D_3 = dose which would be given by $(Q_I')_0$ if $\lambda = \text{constant} = \lambda_m$

$$K = \frac{RM/Q_S + k_2 + k_3}{\lambda_m + k_2 + k_3}$$

It is shown in Appendix II that

$$D < D_1 + D_2 + D_3/K.$$

If m is the initial amount of iodine in the I compartment and is changed at $t = 0$ to M (by the administration of stable iodine), λ_m is necessarily smaller than R, and M could, if necessary, be made large compared with Q_S. The interest lies in finding how large K can be made by practicable increase of M. D_2 decreases as M increases. Hence, the larger the value of M, the smaller will be the upper limit to the thyroid dose.

The above formula provides an example of a conclusion which appears intuitively correct but contains a factor which cannot be estimated without making use of some assumption about the way in which uptake varies with the amount of available iodine. It seems obvious that the dose will at least include the dose (D_1) due to the active iodine originally in the thyroid, that it will exceed this dose at least by the dose (D_2) which would be given if λ had its final value, and that it will be increased by some fraction of the dose (D_3) which would be given if λ had retained its initial value. The difficulty lies in assessing the numerical values since direct evidence on some of the constants is not available. In the following section suggestions are put forward for numerical values which appear to be consistent with the general features revealed by measurements in cases of iodine excess or deficiency.

VII. NUMERICAL VALUES

RIGGS[8] quotes "illustrative values" for many of the quantities involved in the iodine regime. The relation between the rate constants to which numbers are assigned and the rate constants used in the foregoing theory are considered below. The terminology and definitions used by RIGGS[8] are given in quotes.

"K_{IG} rate constant of iodine uptake by the thyroid gland"

K_{IG} is thus identical with k_1 in an equilibrium state

"K_{IE} rate constant of renal excretion of iodide"

K_{IE} should thus be taken as $g\,k_2$, but the implication is that RIGGS takes g to be unity and hence $k_{IE} \simeq k_2$

"K_{GB} rate constant of secretion of organic iodide from thyroid"

K_{GB} is thus identical with k_4

"K_G rate constant of net loss of radioactive iodine from thyroid sufficiently long after a tracer dose so that the ratio of specific activity in G to specific activity in B is constant, rate of biological decay in G"

(The implication of this definition is considered later in this section)

"K_{BI} rate constant of breakdown of hormone in B to inorganic iodine"

K_{BI} is thus identical with k_6

"K_{BF} rate constant of excretion of organic iodine"

If only urinary excretion is considered, K_{BF} should be taken as fk_5. If total excretion is considered, K_{BF} is identical

with k_5. It is assumed below that f is sufficiently close to unity to justify the assumption $K_{BF} = k_5$.

The numerical values in Table 4 below are quoted from Table 3 of Ref. 8. The value of k_3

Table 4.

Quantity	"Normal value" (hr^{-1})	Value in chronic iodine deficiency (hr^{-1})
$k_1 = K_{IG}$	0.0389	0.622
$gk_2 = K_{IE} \simeq k_2$	0.0800	0.0800
$k_4 = K_{GB}$	0.000364	0.000972
$k_5 = K_{BF}$	0.000208	0.000208
$k_6 = K_{BI}$	0.00222	0.00222

is taken as $k_3 = 0.0036\ hr^{-1}$, corresponding with a half-life of approximately 8 days.

The increase in $k_1 = K_{IG}$ from 0.0389 in the normal case to 0.622 in the case of chronic deficiency is a change in a direction which accords with the hypothesis which has been put forward. The fact that $k_4 = K_{GB}$ shows a smaller increase (a factor of less than 3 against a factor of about 16) suggests that k_4 is a slowly varying function of Q_G. It is, however, proposed to disregard this variation in the calculation of total dose, on the following grounds: (a) k_4 occurs in the expression for the total dose only in association with k_3, i.e. in a coefficient $(k_3 + k_4)$. A reduction of k_4 below the value 3.6×10^{-4} would make little difference to the value of $k_3 + k_4$ and hence the effect on the total dose can be neglected; (b) the analysis can be regarded as applying to a case in which a variable k_4 is replaced by its appropriate mean value. In the application to total dose calculations the "reduction factor"

$$\frac{(\lambda_m + k_2 + k_3)}{(RM/Q_S + k_2 + k_3)}$$

is independent of k_4.

To interpret the meaning of K_G it is necessary to refer to the analysis given in Appendix II. It is there shown that the model leads to an expression for the quantity of radioactive iodine in the thyroid which is the sum of three exponentials. The exponents are shown to be roots of an equation [equation (10)] in which the coefficients of the terms are determined by the k's. Thus when the k's are known the exponents are known. If the numerical values of the exponents differ fairly considerably from one another there will thus be three regions of the experimental curves such that, within each region, the curve will approximate to a simple exponential whose rate constant is given as one of the solutions of equation (10). POCHIN[11] has referred to the interpretation of an empirical curve, plotting thyroidal radioactive iodine content against time, in terms of three intervals. He points out that the slopes of the curve can be regarded as comprising an uptake period, a period governed mainly by the secretion rate and, at relatively long times, by a period in which the slope is mainly determined by the rate of recycling. For the particular case quoted by POCHIN the thyroid was "overactive", giving unusually high uptake and secretion rates. In other cases there would be a different correspondence between rate constants and successive parts of the relevant curve. Nevertheless three intervals will, on the assumptions of the model, be recognisable on the curve provided the roots of equation (10) differ sufficiently amongst themselves.

The definition of K_G implies that it refers to the rate constant which is effective at very long times, i.e. at times when the terms involving the other rate constants can be regarded as negligible. The quoted numerical value of K_G is 0.000255. Hence if this interpretation is correct $\mu = -0.000255$ should be an approximate solution of equation (10). Inserting the values of the k's it is clear that this value of μ is an approximate solution of the equation. Since its numerical value is also an order or more smaller than those of the other two approximate roots $(k_1 + k_2$ and $k_5 + k_6)$ there will be a late time range in which the decay in G will apparently be governed by K_G.

It is clear that K_G is not an independent constant, but is defined in terms of the other K's if the rigorous solutions of the equations are used. Hence to the extent that calculated and observed values of K_G agree, the three-compartment model is justified as a mathematical basis.

If the formula is to be used to estimate dose reduction it is necessary to give numerical values

to R and Q_S. One method of obtaining estimates of these parameters is to assume that the formula, $\lambda = R(1 - Q_G/Q_S)$ applies over the whole range of values of Q_G from "normal" to "chronic iodine deficiency". The numerical values quoted by RIGGS are:

	Normal value	Value in chronic iodine deficiency
λ	0.0389 hr^{-1}	0.622
Q_G	8 mg	3 mg

Substitution of these values in the formula gives $R = 0.973\ hr^{-1}\ Q_S = 8.34$ mg, i.e. $R/Q_S \simeq 0.117$.

For present purposes it is necessary to consider applications only for values of Q_G at and above the "normal" value of 8 mg. With this objective in mind it is desirable to examine the possible range of the constants without using the values quoted for iodine deficiency.

One numerical relation between R and Q_S in this range follows at once if the formula is fitted to the illustrative values quoted above for the "normal" case. This is:

$$0.0389 = R(1 - 8/Q_S)$$

One further numerical postulate is thus necessary to determine R and Q_S separately. There are difficulties in finding evidence for such a further relation. One difficulty arises because appropriate measurements have not, so far as the authors are aware, been reported for an "equilibrium" condition in which the intake is maintained at a value well above "normal" for "normal" patients. A second difficulty is that some of the evidence, e.g. graphs of Fig. 2 referring to the thyroid, indicate that, at least in the case of a large single dose of total iodine administered to patients with exophthalmic goitre, the initial stages are more complex than those assumed in the model. The initial stages to which reference is made here are not those concerned with the rate of absorption of the iodine by the bloodstream. It is true that any observations made within an hour or two may require an interpretation which takes this process into account. However, in the present context it appears legitimate to neglect this period, which is very brief compared with the period which is effective in the accumulation of radioactive dose. After the maximum uptake has occurred, the thyroid curves referring to large doses of total iodine appear to indicate a period during which the reduction of activity occurs at a rate which is considerably higher than that occurring when the accompanying stable iodide is 0.1 mg or less. In each of the 10-mg curves of Fig. 2, the "uptake" levels off to about 5 per cent or less, indicating a temporary increase in the thyroidal iodine content of 0.5 mg or less. However the very short "overshoot" indicates an increase of 3 mg. It might be arguable that this level would tend to have been maintained if the available iodine had been kept at 10 mg by continued administration of iodine to balance the loss by secretion. If so, it would be necessary to assume that these particular patients could accommodate at least 3 mg of iodine in excess of that normally present. What percentage this is of their "normal" thyroidal iodine content cannot be determined. It may be surmized that, since the uptake can be taken approximately as proportional to $1/(1 + k_2/k_1)$ and the uptakes shown are in the region of $\frac{2}{3}$, k_1 was about double k_2. The "normal" value for k_1 is about half k_2. Since the thyroid iodine mass can, in the terms of the model, be taken as approximately proportional to k_1, it may be that 3 mg in the cases under discussion would correspond only with about $\frac{3}{4}$ mg in "normal" cases. This very speculative argument is introduced only to indicate that the results illustrated in Fig. 2 could be consistent with a "saturation" value only about 10 per cent higher than the normal iodine content even for the patients to whom the results refer.

A conclusion which is indicated with rather more certainty is that the model adopted is oversimplified in relation to the period up to about 1 day after the administration of total doses of iodine of the order 10 mg or more. It appears, as suggested by STANBURY *et al.*,[7] that if the initial effects are to be explained it would be necessary to elaborate the model by introducing another thyroidal compartment. Such elaboration does not seem justified in the present connexion. The additional dose due to the attainment of a temporary peak at about 30 per cent

as against a very much longer sustained level of about 5 per cent will affect the calculated dose by a factor less than 2. Although the phenomenon is of interest, the practical importance is not great in the application considered here.

It appears therefore that no specific figure can be deduced for Q_S (or R). However it is of interest to examine the assumption that a "saturation value" exists for thyroidal iodine, without taking account of the enhanced rate of loss which appears to be associated with massive doses. The calculation can then be put in a form which underestimates the radioactive dose to the thyroid.

The quantity K has been defined as

$$K \simeq \frac{RM/Q_S + k_2 + k_3}{\lambda_m + k_2 + k_3}$$

and the "normal" values when inserted in the formula

$$\lambda = R(1 - Q_G/Q_S)$$

give the relation $R/Q_S = 0.0389/Q_S - 8$ where Q_S is in mg. Numerically K is thus given as

$$K \simeq \frac{0.317\,M}{Q_S - 8} + 0.682$$

The value of Q_S which would be deduced if the formula for λ covered the whole range from gross deficiency to gross excess would, as shown above, be $Q_S = 8.34$ mg. With this value, the equation for K would be

$$K \simeq 0.932\,M + 0.682$$

Thus for values of M of about 10 mg, K would be about 10, and for greater values of M, K would increase about in proportion to M.

However since it cannot be assumed that the formula covers the whole range it is perhaps more useful to consider the values of K for a range of values of Q_S. In the following table this range is arbitrarily taken to extend to 12 mg, i.e. to a "saturation value" for thyroidal iodine content 50 per cent greater than "normal".

Dependence of K on Q_S and M

Q_S	8.34	9.0	10.0	11.0	12.0
Values of K					
M = 10 mg	9.7	3.8	2.3	1.7	1.5
M = 100 mg	91	32	16	11	8.7

No positive evidence can at present be offered on the range in which Q_S lies. It must remain a surmise that the thyroid is likely to be incapable of increasing its iodine content by more than 50 per cent. It is however apparent from the formula for K that even a 100 per cent increase (to 16 mg) would still give a value of K greater than 4.6 for a value of M of 100 mg.

It therefore appears reasonable to the authors to assume that 100 mg of stable iodine will give a dose reduction by a factor of about 10 (the approximate value observed in their own cases) and which is very unlikely to be less than 5. The administration of quantities of stable iodine of the order of 100 mg is considered in the next section in relation to the possibility of toxic or allergic reactions.

VIII. THERAPEUTIC ASPECTS AND TOXIC REACTIONS OF IODIDE AND ANTITHYROID DRUGS

The incidence of toxic or allergic reactions to iodine and iodides has been assessed by a search of the literature. Elemental iodine is more toxic than iodides, but the belief that iodine is highly toxic is a popular fallacy.[12] Fatal reactions are extremely rare and are the result of either an overwhelming intoxication following large doses of iodine, or bullous ioderma which occasionally occurs in patients who have been taking iodides over long periods.[13] Eighteen fatal cases had been reported by 1942.[14] Few patients who take iodine with suicidal intent are successful in their attempts. Of a series of 1195 cases of attempted suicide treated at the Boston City Hospital between 1915 and 1936, 327 ingested iodine in some form; no death was recorded in this large group.[15] Eighteen deaths were, however, reported from a study of the records of the Medical Examiner's Office of New York City in the six years 1931–37.[16] In all these cases death was due to the local corrosive action of iodine on the oesophagus, stomach or duodenum following ingestion of tincture of iodine. An assessment of the dose had been made in only nine cases and in one case was only 1 oz (fluid). Strong Iodine Solution (U.S.P.) contains 5 per cent w/v I_2 and 10 per cent potassium iodide. The fatal dose of iodine in one case could therefore have been

1.6 g iodine and 3.2 g potassium iodide, which confirms the view that elemental iodine is more toxic than iodides.[12]

Acute poisoning from an initial dose of iodide is relatively rare and even after intravenous injections reactions are seldom seen.[12] The reported cases of poisoning by iodide are almost invariably the result of repeated iodide administration, particularly for the treatment of hyperthyroidism and in antisyphilitic therapy,[17,18,14] or in patients suffering from advanced renal failure or cardiac failure.[19–21] Iodides are widely used as expectorants. Mist. Potassium Iodide (B.N.F.)[22] is one of the more popular expectorant mixtures; reactions to it are extremely rare. SWINNEY[23] reported fifty-four cases of toxic reactions in 1100 patients on oral potassium iodide therapy. None of these cases was serious and the symptoms subsided on withdrawal of the drug. In his review of iodide therapy in syphilis SNODGRASS[17] states that iodism occurred more frequently when the initial dose of iodide was 5 grains or less (12 per cent of cases) than when the initial dose was more than 30 grains (1 per cent of cases), but this refers to effects in a prolonged period of iodine therapy. There are no reported cases of iodism following the ingestion of single small doses of iodide such as is envisaged here.

Patients suffering from chronic renal and cardiac failure are known to be hypersensitive to iodides and iodine, and fatal reactions have been described in patients suffering from these diseases following prolonged iodine therapy. Iodides are also said to cause irritative reactions in tuberculous patients and may even activate a dormant lesion. Although iodide therapy is therefore contra-indicated in persons known to be suffering from pulmonary tuberculosis,[12] small single doses would not lead to any untoward effects.

As defined by ASTWOOD and SOLOMON,[24] the term "antithyroid" drug is applied to a group of compounds which interfere with the incorporation of inorganic iodide by the thyroid gland into the organic precursors of the thyroid hormones. Of this group of drugs the most commonly used in the treatment of hyperthyroidism are derivatives of thiourea (thiouracil, methyl and propyl thiouracil, methimazole and carbimazole). These substances prevent the oxidation of iodide in the thyroid gland. This reaction is essential for the first stage of organic binding of iodine to form the specific thyroid hormones. Administration of therapeutically effective doses of these compounds prevents the accumulation of iodine in the thyroid. The gland continues to utilize the organic iodide already present and hence there is a delay before there is symptomatic evidence of interference with normal thyroid function.

MACGREGOR and MILLER[25] demonstrated the complete inhibition of radioiodine uptake by the thyroid gland of normal persons ½–1 hr after the administration of carbimazole. The recommended therapeutic dose of this drug is 10 mg. Propylthiouracil in a dose of 100 mg produces a similar effect which lasts for 3–4 hr.[26]

A second group of chemical substances interferes with the iodine-concentrating mechanism of the thyroid; the best known of these are potassium thiocyanate and potassium chlorate. These substances inhibit the selective absorption by the thyroid of inorganic iodide from the plasma. They are ineffective in the presence of high-blood inorganic iodide levels. Thiocynate is a toxic material and its use could not therefore be contemplated. Perchlorate, on the other hand, is both effective and less toxic than the thiourea derivatives in the treatment of thyrotoxicosis.[27,28] The action of these substances on the thyroid is reversed by the administration of iodide.

Irradiation of the thyroid occurring as the result of inhalation of radioactive iodine can therefore be reduced by either the administration of a single dose of iodide, e.g. 5–10 grains (300–600 mg) of potassium iodide, or the administration of antithyroid drugs.

The toxicity of iodide is negligible, whereas the antithyroid drugs are potential bone-marrow depressants; leucopenia occurs in an appreciable number of cases of hyperthyroidism treated by this means. In addition, drug rashes, arthalgia and pyrexia have been reported during the administration of these substances, although carbimazole is less toxic than other members of this group.[29] Since children are more susceptible to radiation damage than adults,[5] any procedure aimed at protecting the general public must be efficacious in children and young adults.

Potassium iodide could be administered in suitable doses to children with complete safety, whereas experience with the antithyroid drugs in children is not available, and their widespread application in the present state of knowledge would not therefore be acceptable. The administration of inorganic iodide to unselected groups of the general public is therefore preferred to the antithyroid drugs. The reported cases of serious or fatal reactions to iodide or iodine have occurred during the course of prolonged medication and there is no evidence to suggest that untoward reactions would be expected to arise following the administration of 300–600 mg of iodide by mouth to unselected groups of the general population.

OLINER *et al.*[30] studied the iodine uptake of the thyroid and the serum protein-bound iodine-131 (PBI-131) values in eighty-three children whose ages ranged from $2\frac{1}{2}$ months to 18 years. Of these, sixty-one were considered normal as far as their thyroid function was concerned and twelve had various thyroid disorders. Children up to the age of 10 years were given 22–25 μc I-131 and those over 10 years were given 50 μc as a test dose.

Results of the uptake studies on these sixty-one children were compared with sixty-four euthyroid adults investigated by the authors by the same technique. The range of the uptake in the children was 17–50 per cent, with a mean of 31.1 ± 7.63; for adults the range was 16–50 per cent with a mean of 32.17 ± 7.26.

A comparison of the serum PBI-131 levels in the different age groups was, however, highly significant.

The serum PBI-131 level expressed as percentage of dose per litre at 24 hr was:

euthyroid adults	0.0298 ± 0.015
children 10–18 years	0.031 ± 0.02
children 5–9 years	0.049 ± 0.028
children 1–4 years	0.125 ± 0.07

These results indicate that up to the age of 4 years there is a state of mild thyroid hyperactivity, gradually diminishing up to the age of 9 years.

Recommended dose of inorganic iodide

Following consideration of all the available relevant data regarding iodine metabolism and the toxic effects of iodine and iodides taken by mouth both in children and in adults, the authors are convinced that the administration of iodide is both a safe and effective method of reducing irradiation of the thyroid gland in the circumstances discussed earlier. Iodides are commonly prescribed for adults in the familiar Mist. Pot. Iod. Ammon.;[22] each $\frac{1}{2}$ oz (fluid) of this mixture contains $2\frac{1}{2}$ grains (170 mg) of potassium iodide; the mixture is prescribed at a dose of a $\frac{1}{2}$ oz (fluid) three times daily. For children, a commonly prescribed mixture is *Mist. Belladonna et Ephedrine Pro Infans*[22] which contains 1 grain (70 mg) potassium iodide to each 60 minims; again the prescribed dose is 60 minims three times daily. It is quite certain that these quantities of potassium iodide can be prescribed to unselected groups of individuals without fear of any untoward side effects.

It is, therefore, recommended that the iodide should be administered to adults in tablet form, each tablet containing 100 mg of potassium iodide, to be taken by mouth dissolved in milk or water, this dose to be repeated in 4 hr. Similarly, it is recommended that children up to the age of 4 years should be given tablets of 60 mg potassium iodide by mouth, dissolved in milk or water, again the dose to be repeated in 4 hr.

In certain circumstances administration of the antithyroid drugs would be preferred to iodides. In particular, men employed at a nuclear reactor at the time of an accident would have been exposed to a higher concentration of radioactive iodine. Suitable doses of one of these substances, preferably carbimazole (neo-mercazole), should be administered under the supervision of the nursing staff employed on the site. Furthermore, men who could become exposed to high concentrations of iodine vapour during reparative procedures to the plant could be protected by the administration of this drug prior to the commencement of the specified work. Since the total number involved would be small and these men would be employed on the site, careful medical supervision would be available.

IX. APPLICATION TO EMERGENCY SCHEMES AT NUCLEAR POWER STATIONS

From the foregoing it is concluded that it would be safe to administer a quantity of stable iodine whose effect would be to reduce the radiation dose from radioactive iodine concurrently or subsequently inhaled to an estimated value of one tenth of that which would otherwise be delivered to the thyroid. In considering the application to accident conditions it is necessary to take account of the amount of radioactive iodine inhaled before the stable iodine is administered. If a radioactive release occurs from a reactor it will be necessary to estimate iodine concentrations in the downwind direction by means of air samples, to assess meteorological factors, to forecast from the plant conditions and the remedial action at the site the likely duration of the release, and to mobilize vehicles which may be required for evacuation. Let T be the period occupied by this action. T is thus a period during which radioactive iodine is being inhaled both in the case of evacuation and in the case of administration of stable iodine. For simplicity it will be assumed that throughout the whole release an individual in a given position is accumulating radioactive iodine in his I compartment, through inhalation, at the rate of q units/hr. After time T the amount inhaled is qT and the amount in the thyroid is approximately $\frac{1}{2} k_1 q T^2$. In the case of evacuation, with no administration of stable iodine, the radiation dose to the thyroid resulting from an inhalation qT, and hence a thyroidal uptake of approximately $k_1 qT/(k_1 + k_2)$, is proportional to I where

$$I \simeq \frac{k_1 q T}{k_1 + k_2} \cdot \frac{1}{k_3 + k_4}$$

If stable iodine is administered at time T, the dose D_1 due to the radioactive iodine already in the thyroid (of amount $\frac{1}{2} k_1 q T^2$), is given by

$$D_1 = \tfrac{1}{2} k_1 q T^2 \cdot \frac{1}{k_3 + k_4} = \frac{T}{2} (k_1 + k_2) I$$

An analysis of the action which would be taken in emergency conditions at a nuclear power station suggests that it is realistic to take a value of about 1 hr for T. The "normal" value for $(k_1 + k_2)$ can be taken as about 0.12, giving

$$D_1 \simeq 0.06 I$$

If the quantity M of stable iodine is 100 mg or more, it is an over-estimate to take D_2 as high as 0.1 D_3. Thus with 10 per cent each allowed for D_1 and D_2 and with a factor 10 assigned to K, a further 8 hr of inhalation at the rate q could be allowed before the thyroid dose would equal that corresponding with evacuation at 1 hr, with no administration of stable iodine.

The above numerical values are quoted only to illustrate a type of condition which might be met in practice. If a serious release of radioactivity occurred it is envisaged that any individuals in close proximity to the station in the downwind direction would be evacuated in any case. Due heed would be given to hazards other than inhalation before these individuals would be allowed to return. It is suggested that even for such individuals a worthwhile saving in thyroid dose could be effected by giving them stable iodide. The analysis given in this paper is however considered to be more applicable to action beyond this short evacuation range. Lag time is of first importance in this aspect. The discussion has therefore been directed towards providing an estimate of the relation between dose reduction and the time interval before the stable iodide is administered. In the marginal areas beyond the immediate evacuation distance there must be doubt whether evacuation will be required. Some estimate will be required of the time during which decision can be deferred. It is the object of this paper to provide a basis for extending this time with safety so that action can depend on actual measurements of air concentration over several hours rather than on forecasts. There is no suggestion that the arithmetic should be used to justify a dose closely approaching the permissible limits.

In conclusion, the authors should emphasise that, although the merits of the proposals have in their view been substantiated by the arguments given in the paper, they are well aware that conservative estimates of the gain should be used until more information is available on the range of the parameters.

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APPENDIX I*

Blockage of Normal Thyroid Function using 100 mg KI

Measurements of the uptake of I^{131} by the thyroids of the two normal volunteers were made using a multiple-counter technique. I^{131} was used in preference to I^{132} as it was thought necessary to study the degree of retention of the isotope by the thyroid over at least 48 hr, particularly when it was given together with a large amount of carrier iodide. Five microcuries of I^{131} was given orally and counts were made 6, 24, 30 and 48 hr after administration. The test was repeated a month later with a further 5 μc of I^{131}, together with 100 mg of KI.

The counting arrangement[31] consists of four thallium-activated NaI crystals, each 1 in. in dia. and 1 in. long, which are arranged in a ring so that the opposing faces of each pair of crystals are 25 cm apart. The crystals are shielded with lead shields 1 in. thick which are mounted flush with the face so that when centred on the thyroid each crystal "sees" a volume of tissue which extends for rather more than 12.5 cm above and below the level of the thyroid. This volume of tissue includes a proportion of the circulating extrathyroidal radioactivity, which will depend on the

* This appendix was contributed by HELEN E. A. FARRAN, New End Hospital, Hampstead, London.

size and shape of the patient. In the average adult, it has been found experimentally, that, when the isotope is given intravenously, the counting rate due to extrathyroidal activity is of the order of 80–90 counts/sec for each microcurie administered. The efficiency of the counters is such that a central source of 1 μc of I^{131} gives a counting rate of the order of 550 counts/sec. Slight variations (480–620 counts/sec) occur in this value owing to several causes, and a standard equal to $\frac{1}{3}$ of the administered activity was always counted before and after each measurement. (See Table I.1.)

Table I.1

Time (hr)	Subject *A*				Subject *B*			
	Tracer		Tracer–KI		Tracer		Tracer–KI	
	Standard (counts/sec)	Neck (counts/sec)	Standard (counts/sec)	Neck (counts/sec)	Standard (counts/sec)	Neck (counts/sec)	Standard (counts/sec)	Neck (counts/sec)
0	—	—	—	40	—	—	—	50
6	970	465	950	220–40	825	715	780	225–50
24	990	935	1000	118–36	895	1055	780	135–45
30	720	740	830	95–35	770	880	735	115–45
48	870	880	800	110–32	—	—	685	130–40
54	680	700	—	—	—	—	—	—
96	—	—	—	—	640	735	—	—

In the euthyroid subject, under normal conditions, the counting rate due to extrathyroidal activity will rise to a maximum within an hour of giving an oral dose[32] and will then fall again owing to clearance of the iodide from the plasma by the thyroid and kidneys and also because of diffusion of the iodide into the tissues.

In the case of Subject *A*, if we assume that the counting rate of 935 counts/sec measured after 24 hr is due entirely to activity concentrated in the thyroid, then the percentage of the radioactive iodine given which is taken up by the thyroid in 24 hr is

$$\frac{935 \times 100}{990 \times 3} = 31 \text{ per cent}$$

where 990 counts/sec is the counting rate due to the standard which is $\frac{1}{3}$ of the administered activity.

In the same subject, 24 hr after receiving a further 5 μc together with 100 mg of KI the counting rate measured 118 counts/sec compared with a standard count of 875 counts/sec. The residual activity still present in the neck after the previous investigation accounts for 38 counts/sec so that, even if all the activity "seen" is concentrated in the thyroid, which is unlikely, there is only 118–38 counts/sec or

$$\frac{82 \times 100}{875 \times 3} = 3 \text{ per cent}$$

of the given activity remaining in the neck.

In both these volunteers it would seem that the administration of 100 mg of KI has reduced the uptake of radioactive iodine to less than 3 per cent of the amount ingested. This finding confirms the observations of Hamilton[33] who found that, when normal subjects were given 14 mg of NaI labelled with I^{131} the thyroid only took up 1–5 per cent of the administered activity.

APPENDIX II*

Solution of the Equations for the Three-Compartment Model

A. The equations for the model

The model adopted is essentially that used by STANLEY *et al.* The symbols are defined in Section III and the model is shown diagrammatically in Fig. 1.

The equations governing the Q's and the Q^x's are

$$\frac{dQ_I}{dt} = -(k_1 + k_2)Q_I + k_6 Q_B + I_n \qquad (1)$$

$$\frac{dQ_G}{dt} = -k_4 Q_G + k_1 Q_I \qquad (2)$$

$$\frac{dQ_B}{dt} = -(k_5 + k_6)Q_B + k_4 Q_G \qquad (3)$$

$$\frac{dQ_I^x}{dt} = -(k_1 + k_2)Q_I^x + k_6 Q_B^x \quad (4)$$

$$\frac{dQ_G^x}{dt} = -k_4 Q_G^x + k_1 Q_I^x \quad (5)$$

$$\frac{dQ_B^x}{dt} = -(k_5 + k_6)Q_B^x + k_4 Q_G^x \quad (6)$$

Solving (4), (5) and (6) by standard methods, the expressions for Q_I^x, Q_B^x, Q_G^x are

$$Q_G^x = \sum_{r=1}^{3} A_r e^{\mu_r t} \quad (7)$$

$$Q_I^x = \frac{1}{k_1} \sum_{r=1}^{3} A_r(\mu_r - \mu_b) e^{\mu_r t} \quad (8)$$

$$Q_B^x = \frac{1}{k_1 k_6} \sum_{r=1}^{3} A_r(\mu_r - \mu_a)(\mu_r - \mu_b) e^{\mu_r t} \quad (9)$$

where μ_1, μ_2, μ_3 are the roots of

$$(\mu + k_1 + k_2)(\mu + k_4)(\mu + k_5 + k_6) = k_1 k_4 k_6 \quad (10)$$

and μ_a, μ_b, μ_c are the corresponding roots of

$$(\mu + k_1 + k_2)(\mu + k_4)(\mu + k_5 + k_6) = 0 \quad (11)$$

i.e.

$$\mu_a = -(k_1 + k_2),\ \mu_b = -k_4,\ \mu_c = -(k_5 + k_6) \quad (12)$$

The values of the A's are given by

$$A_1 = K(\mu_1 - \mu_c)(\mu_2 - \mu_3) \quad (13)$$

$$A_2 = K(\mu_2 - \mu_c)(\mu_3 - \mu_1) \quad (14)$$

$$A_3 = K(\mu_3 - \mu_c)(\mu_1 - \mu_2) \quad (15)$$

where

$$K[\mu_1^2(\mu_2 - \mu_3) + \mu_2^2(\mu_3 - \mu_1) + \mu_3^2(\mu_1 - \mu_2)] = k_1(Q_I^x)_0 \quad (16)$$

and $(Q_I^x)_0$ is the value of Q_I^x at $t = 0$, i.e. the administered dose.

The above rigorous conclusions are clearly consistent with the approximate formulae frequently quoted for cases in which μ_1, μ_2 μ_3 differ from one another by large factors. Thus if μ_2 and μ_3 are each considerably smaller than μ_1 the approximate form for Q_G^x can readily be deduced as applying at early stages

$$Q_G^x \simeq \frac{k_1(Q_I^x)_0}{k_1 + k_2}(1 - e^{-(k_1+k_2)t}) \quad (17)$$

There will similarly be later ranges of t in which the rate of change of Q_G^x will be dominated by one of the other two exponentials.

If $k_1 k_4 k_6$ is sufficiently small the following approximations must clearly be valid:

$$\mu_1 \simeq \mu_a = -(k_1 + k_2),\quad \mu_2 \simeq \mu_b = -k_4,$$
$$\mu_3 \simeq \mu_c = -(k_5 + k_6)$$

More accurate approximations are however directly obtainable in the form

$$\mu_1 = \mu_a + \delta_a,\quad \mu_2 = \mu_b + \delta_b,\quad \mu_3 = \mu_c + \delta_c \quad (18)$$

where

$$\delta_a = \frac{k_1 k_4 k_6}{(k_1 + k_2 - k_4)(k_1 + k_2 - k_5 - k_6)} \quad (19)$$

$$\delta_b = \frac{k_1 k_4 k_6}{(k_1 + k_2 - k_4)(k_5 + k_6 - k_4)} \quad (20)$$

$$\delta_c = \frac{k_1 k_4 k_6}{(k_1 + k_2 - k_5 - k_6)(k_4 - k_5 - k_6)} \quad (21)$$

It follows from (19), (20) and (21) that if k_6 is not small compared with the other k's any analysis of experimental curves should take account of the following:

μ_1—μ_1 can be taken as $\mu_1 = \mu_a = -(k_1 + k_2)$ if k_4 and k_6 are small compared with k_1 or k_2.

μ_2—μ_2 can be taken as $\mu_2 = \mu_b = -k_4$ only if $k_1 k_6$ is small compared with

$$|(k_1 + k_2 - k_4)(k_5 + k_6 - k_4)|$$

μ_3—μ_3 cannot be taken as $\mu_3 = \mu_c = -(k_5 + k_6)$ unless $k_1 k_4 k_6$ is small compared with

$$|(k_1 + k_2 - k_5 - k_6)(k_4 - k_5 - k_6)(k_5 + k_6)|$$

It is of interest that the model implies that, in any equilibrium state, a knowledge of any one of the Q's, or I_n (or the total excretion rate) enables the masses of iodine in the other compartments to be deduced. Thus equations (1), (2) and (3) show that in an equilibrium

$$Q_I = \frac{k_4}{k_1} Q_G \quad (22)$$

$$Q_B = \frac{k_4}{k_5 + k_6} Q_G \quad (23)$$

$$I_n = \left[(k_1 + k_2)\frac{k_4}{k_1} - \frac{k_4 k_6}{k_5 + k_6}\right] Q_G \quad (24)$$

B. Accumulation of iodine in urine

The constants f, g, P, β as defined in Table 3 are not independent. Any pair can be assigned values between 0 and 1, and the values for the other pair will then be determined. From the definitions it

follows that g and f can be expressed in terms of P and β as follows

$$g = P(1 - \beta)\left[\frac{k_5}{k_5 + k_6} \cdot \frac{k_1}{k_2} + 1\right] \quad (25)$$

$$f = \frac{P\beta g}{g - P(1 - \beta)} \quad (26)$$

The equation for the rate of increase of radioactive iodine in the urine is most conveniently expressed by using f and g as the parameters and is

$$\frac{dQ_u{}^x}{dt} = f\,k_5 Q_B{}^x + g\,k_2 Q_I{}^x \quad (27)$$

The solution is

$$Q_u{}^x = P(Q_I{}^x)_0 + \Sigma \frac{A_1}{\mu_1}(\mu_1 - \mu_B)\left[\frac{fk_5}{k_1 k_6}(\mu_1 - \mu_a) + g\frac{k_2}{k_1}\right]e^{\mu_1 t} \quad (28)$$

where the summation Σ applies to the three values of μ and A. For the normal case, in which $\mu_1 - \mu_a$ is numerically small compared with k_1, k_5 small compared with k_6 and μ_b small compared with μ_1, the solution for the initial period during which $e^{\mu_3 t}$ and $e^{\mu_2 t}$ may each be regarded as unity can be simplified to

$$Q_u{}^x \simeq \frac{gk_2}{k_1 + k_2}(Q_I{}^x)_0[1 - e^{-(k_1 + k_2)t}] \quad (29)$$

C. Dose to the thyroid when the rate constants do not vary

If D is the total dose to the thyroid, D is proportional to I where

$$I = \int_0^\infty Q_a{}^1\,dt = \int_0^\infty \Sigma\, A_1 e^{(\mu_1 - k_3)t}\,dt$$

Hence if $\mu^1 = \mu - k_3$, $\quad I = -\Sigma \dfrac{A_1}{\mu_1{}^1}$.

Using the fact that $\mu_1{}^1$, $\mu_2{}^1$, $\mu_3{}^1$ are the roots of

$$(\mu^1 - \mu_a{}^1)(\mu^1 - \mu_b{}^1)(\mu^1 - \mu_c{}^1) - k_1 k_4 k_6 = 0,$$

I can be derived as

$$I = \frac{k_1 \mu_c{}^1 (Q_I{}^x)_0}{\mu_a{}^1 \mu_b{}^1 \mu_c{}^1 + k_1 k_4 k_6} \quad (30)$$

(30) gives I explicitly in terms of the k's. For probable relative magnitudes of the latter, (30) can be approximated to

$$I \simeq \frac{k_1 (Q_I{}^x)_0}{(k_1 + k_2 + k_3)(k_3 + k_4)} \quad (31)$$

It is clear from (31) that, where the approximation is valid, I diminishes as k_1 diminishes. The same result can readily be demonstrated from (30). Thus any action which decreases the value of k_1 will decrease the dose to the thyroid.

In following sections, consideration is given to the case in which k_1 is not a constant but a function of Q_G. The analysis is simplified if the recycling of iodine can be neglected, i.e. if k_6 can be taken as zero. The object of the analysis is to obtain an estimate of the dose reduction which can be achieved by creating conditions which lead to a reduction of k_1. It is therefore useful to find whether a conservative estimate of the reduction can be made by treating the case in which $k_6 = 0$.

For this purpose I can be regarded as a function of two parameters only, k_1 and k_6, since the other parameters will remain constant. Two possible values of k_1 will be considered, k_1 and $k_1{}^1$ where $k_1{}^1 < k_1$, and two values of k_6, i.e. k_6 and 0.

The true ratio of the dose corresponding with k_1 to that corresponding with $k_1{}^1$ is $I(k_1,k_6)/I(k_6{}^1,k_6)$.

The ratio assessed on the basis $k_6 = 0$ is $I(k_1, 0)/I(k_1{}^1, 0)$. $I(k_1 k_6)$ can be written as $I(k_1 k_6) = C/(K_1 - B)$, where

$$C = \frac{(Q_1{}^x)_0}{-\mu_b{}^1}, \quad K_1 = \frac{1}{k_1}(k_1 + k_2 + k_3), \quad B = \frac{k_4 k_6}{\mu_b{}^1 \mu_c{}^1}$$

i.e. C, K_1 and B are all positive.

Hence

$$\frac{I(k_1 k_6)}{I(k_1{}^1 k_6)} = \frac{K_1{}^1 - B}{K_1 - B}$$

where $$K_1{}^1 = \frac{1}{k_1{}^1}(k_1{}^1 + k_2 + k_3)$$

and $$\frac{I(k_1, 0)}{I(k_1{}^1, 0)} = \frac{K_1{}^1}{K_1}$$

$$\frac{K_1{}^1 - B}{K_1 - B} \gtrless \frac{K_1{}^1}{K_1}$$

if $-K_1 B > -K_1{}^1 B$.

i.e. if $K_1 < K_1{}^1$, or

$$1 + \frac{k_2 + k_3}{k_1} < 1 + \frac{k_2 + k_3}{k_1{}^1}$$

i.e. if $k_1{}^1 < k_1$.

Thus if $k_1{}^1 < k_1$ the ratio $I(k_1, 0)/I(k_1{}^1, 0)$ underestimates the ratio $I(k_1, k_6)/I(k_1{}^1, k_6)$. If $k_1{}^1$ varies but is always less than k_1 the above result indicates that the ratio will also be underestimated if the condition $k_6 = 0$ is assumed.

D. Dose to the thyroid when the rate constants vary

The constant k_1 is replaced by a variable parameter λ and it is assumed that in the range of Q_G concerned λ can be expressed as

$$\lambda = R\left(1 - \frac{Q_G}{Q_S}\right) \tag{32}$$

If this linear formula were to apply over the whole range of variation of Q_G, R would be the rate constant for a thyroid totally deprived of iodine, and Q_S would be the limiting mass of iodine, asymptotically approached, which the thyroid could accommodate. It is not necessary to regard R as having the above physical meaning since the linear relationship need be applied only over the range from normal iodine content to considerable iodine excess. The constant Q_G can however be regarded as plausibly representing a limiting iodine mass in the thyroid.

The case which will be considered is that in which, up to time $t = 0$, an equilibrium has been established with an iodine content $Q_I = m$. At $t = 0$ it will be assumed that the value is changed to, and maintained at, $Q_I = M$.

From (2) the equation for Q_G is

$$\frac{dQ_G}{dt} = -k_4 Q_G + \lambda M$$

or, using (32),

$$\frac{dQ_G}{dt} = RM - Q_G\left(k_4 + \frac{RM}{Q_S}\right)$$

Hence if $(Q_G)_0$ is the value of Q_G at $t = 0$,

$$Q_G = \frac{RM}{k_4 + (RM/Q_S)} + \left[(Q_G)_0 - \frac{RM}{k_4 + (RM/Q_S)}\right] e^{-(k_4 + RM/Q)t} \tag{33}$$

The asymptotic value of Q_G is thus

$$\frac{Q_S}{1 + k_4 Q_S/RM} \tag{34}$$

If M is sufficiently large this asymptotic value approximates to Q_S. Let λ_m = rate constant corresponding with an equilibrium in which $Q_I = m$, and λ_M = rate constant corresponding with an equilibrium in which $Q_I = M$.

From (32) and (34)

$$\lambda_M = \frac{R}{1 + RM/k_4 Q_S} \tag{35}$$

and

$$(Q_G)_0 = Q_S - \frac{\lambda_m Q_S}{R} \tag{36}$$

Hence, using (33),

$$\lambda = \lambda_M + (\lambda_m - \lambda_M) e^{-Rk_4 t/\lambda_M} \tag{37}$$

For the conditions assumed, λ is thus known explicitly in terms of t. On the assumption $k_6 = 0$ the equations for Q_I^1 and Q_G^1 are

$$\frac{dQ_I^1}{dt} = -(\lambda + k_2 + k_3) Q_I^1 \tag{38}$$

$$\frac{dQ_G^1}{dt} = -(k_3 + k_4) Q_G^1 + \lambda Q_I^1 \tag{39}$$

$$\text{Let } J = \int_0^t \lambda\, dt = \lambda_M \left[t + \frac{(\lambda_m - \lambda_M)}{Rk_4} \times (1 - e^{-Rk_4 t/\lambda M}) \right] \tag{40}$$

Direct integration of (38) gives

$$Q_I^1 = (Q_I^1)_0\, e^{-[J + (k_2 + k_3)t]} \tag{41}$$

The total dose to the thyroid is proportional to D where

$$D = \int_0^\infty Q_G^1\, dt$$

From (39)

$$D = \int_0^\infty Q_G^1\, dt = \frac{1}{k_3 + k_4}\left[\int_0^\infty \lambda Q_I^1\, dt - \left|Q_G^1\right|_0^\infty\right] \tag{42}$$

The value of Q_G^1 at $t = \infty$ is zero. At $t = 0$ it will be assumed that there is a quantity of radioactive iodine $(Q_G^1)_0$ in the thyroid. Thus from (42) and (41)

$$\frac{(k_3 + k_4)D - (Q_G^1)_0}{(Q_I^1)_0} = \int_0^\infty \lambda e^{-[J + (k_2 + k_3)t]}\, dt$$

Since, by definition, $\frac{dJ}{dt} = \lambda$

$$\frac{(k_3 + k_4)D - (Q_G^1)_0}{(Q_I^1)_0} = -\int_0^\infty \left[\frac{d}{dt} e^{-[J + (k_2 + k_3)t]} + (k_2 + k_3) e^{-[J + (k_2 + k_3)t]}\right] dt$$

$$= 1 - (k_2 + k_3) \int_0^\infty e^{-[J + (k_2 + k_3)t]}\, dt \tag{43}$$

The exponent in (43) is of the form

$$-[a + bt - ae^{-\alpha t}]$$

$$\text{Let } I = \int_0^t e^{-(a + bt - ae^{-\alpha t})} dt$$

and let $ae^{-\alpha t} = u$

Thus $$I = \frac{e^{-a}}{\alpha a^{b/\alpha}} \int_u^a u^{b/\alpha - 1} e^u \, du$$

or $$I = \frac{e^{-a}}{ba^{b/\alpha}} \left[\left| u^{b/\alpha} e^u \right|_u^a - \int_u^a u^{b/\alpha} e^u \, du \right.$$

or $$I = \frac{1}{b} \left(1 - e^{-(a+bt-ae^{-\alpha t})} \right) - \frac{e^{-a}}{ba^{b/\alpha}} \int_u^a u^{b/\alpha} e^u \, du \qquad (44)$$

Thus from (43) and (44)

$$\frac{(k_3 + k_4)D - (Q_a{}^1)_0}{(Q_I{}^1)_0} = 1 - \frac{(k_2 + k_3)}{b} \times \left(1 - \frac{e^{-a}}{a^{b/\alpha}} \int_0^a u^{b/\alpha} e^u \, du \right) \qquad (45)$$

where $a + bt - ae^{-\alpha t} = J + (k_2 + k_3)t$ and hence

$$\left.\begin{aligned} a &= \frac{\lambda_M(\lambda_m - \lambda_M)}{Rk_4} \\ b &= \lambda_M + k_2 + k_3 \\ \alpha &= \frac{Rk_4}{\lambda_M} \end{aligned}\right\} \qquad (46)$$

In the application under consideration $\lambda_m > \lambda_M$ and hence a, b and α are positive.

Hence

$$\frac{e^{-a}}{a^{b/\alpha}} \int_0^a u^{b/\alpha} e^u \, du < \frac{1}{a^{b/\alpha}} \times \int_0^a u^{b/\alpha} = \frac{1}{a^{b/\alpha}} \frac{u^{b/\alpha + 1}}{b/\alpha + 1} \Big|_0^a = \frac{a}{b/\alpha + 1} \qquad (47)$$

Thus if $G = \dfrac{(k_3 + k_4)D - (Q_a{}^1)_0}{(Q_I{}^1)_0}$, from (45) and (47),

$$G < 1 - \frac{(k_2 + k_3)}{\lambda_M + k_2 + k_3} \times \left[1 - \frac{\lambda_m - \lambda_M}{\lambda_M + k_2 + k_3 + (Rk_4/\lambda_M)} \right]$$

i.e.

$$G < \frac{\lambda_M}{\lambda_M + k_2 + k_3} + \frac{\lambda_m}{k_2 + k_3 + (R\,k_4/\lambda_M)}$$

From (35), $$\frac{Rk_4}{\lambda_M} = k_4 + \frac{RM}{Q_S}$$

Hence

$$G < \frac{\lambda_M}{\lambda_M + k_2 + k_3} + \frac{\lambda_m}{(RM/Q_S) + k_2 + k_3}$$

or

$$D < \frac{(Q_I{}^1)_0}{k_3 + k_4} + \frac{(Q_I{}^1)_0}{k_3 + k_4} \left[\frac{\lambda_M}{\lambda_M + k_2 + k_3} + \frac{\lambda_m}{\lambda_m + k_2 + k_3} \cdot \frac{\lambda_m + k_2 + k_3}{RM/Q_S + k_2 + k_3} \right] \qquad (48)$$

The approximation quoted in equation (31), viz.

$$I \simeq \frac{k_1 (Q_I{}^1)_0}{(k_1 + k_2 + k_3)(k_3 + k_4)}$$

is the approximation to a quantity proportional to the radiation dose delivered to the thyroid on the assumption that the k's are constant and $k_6 = 0$.

Thus, on these assumptions, doses D_1, D_2, D_3 can be defined as follows:

D_1 = dose resulting from initial radioactive iodine $(Q_a{}^1)_0$ in thyroid

D_2 = dose resulting from $(Q_I{}^1)_0$ if λ = constant = λ_M

D_3 = dose resulting from $(Q_I{}^1)_0$ if λ = constant = λ_m

and (48) shows that

$$D < D_1 + D_2 + \frac{D_3}{K}$$

where $$K = \frac{(RM/Q_S) + k_2 + k_3}{\lambda_m + k_2 + k_3} \qquad (49)$$

R is necessarily greater than λ_m and M can be made large compared with Q_S. Since increase of M also makes D_2 small compared with D_3, $D - D_1$ can be made small compared with D_3. The dose D_1 is delivered whether or not stable iodine is administered at $t = 0$. Thus a decrease in dose is achievable by administering stable iodine. The magnitude of the reduction depends on the practicable value for M and the relative magnitudes of the other constants. These numerical aspects are discussed in the body of the report.

E. Accumulation of radioactive iodine in urine

It is of interest to consider the equations for accumulation of radioactive iodine in urine when the rate constants vary since results corresponding with high iodine intake have been published in the literature.

On the resumption $k_6 = 0$ the relevant equations are:

$$\frac{dQ_I{}^x}{dt} = -(\lambda + k_2)Q_I{}^x$$

$$\frac{dQ_u{}^x}{dt} = gk_2Q_I{}^x$$

giving $$Q_I^x = (Q_I^x)_0 e^{-(J+k_2)t}$$

and $$Q_u^x = gk_2(Q_I^x)_0 \int_0^t e^{-(J+k_2)t}\, dt$$

The integral is given by (44), using the values for a and $\propto$ defined in (46) and putting $b = \lambda_M + k_2$. Thus

$$\frac{Q_u^x}{g(Q_I^x)_0} \simeq \frac{k_2}{b}\left[1 - e^{-(a+bt-ae^{-\propto t})}\right] - \frac{a\,k_2}{b(b/\propto + 1)}\left|\left(\frac{u}{a}\right)^{b/\propto+1}\right|_u^a.$$

In the applications under consideration λ_M can be considered small compared with k_2, i.e. $b \simeq k_2$. Hence

$$\frac{Q_u^x}{g(Q_I^x)_0} \simeq \left[1 - e^{-(a+bt-ae^{-\propto t})}\right] - \frac{a}{b/\propto + 1}\left[1 - e^{-(b+\propto)t}\right] \quad (50)$$

It is to be noted that (50) is valid as an approximation only for the period for which the error introduced by the assumption $k_6 = 0$ is negligible. It is obvious on physical grounds that the true asymptotic value of $Q_u^x/g(Q_I^x)_0$ is unity in all cases. Approximations for a, b and $\propto$ valid when M is large compared with m (where m is the result of normal dietary intake of iodine) are

$$a = \frac{\lambda_m}{R}\frac{Q_s}{M}, \quad b \simeq k_2, \quad \propto \simeq R\frac{M}{Q_s}$$

The first bracketed term in (50) differs only slightly from an exponential curve characterized by k_2. This result follows since a is small and hence $a(1 - e^{-\propto t})$ starts with the value zero and, because $\propto$ is large, rapidly approaches the small value a. Thus in the absence of the second bracketed term, the urine activity curve would closely resemble an exponential showing Q_u^x approaching $g(Q_I^x)_0$ with a time constant approximating to $1/k_2$.

This behaviour will however be modified by the second bracketed term, whose limiting value is

$$\frac{a}{b/\propto + 1} = \frac{\lambda_m}{k_2 + (RM/Q_s)}$$

i.e. a quantity which decreases as M increases. When M is sufficiently large to make the second term negligible compared with the first, further increase of M cannot produce a detectable effect. The solution is thus consistent with the following observed features of the urine curves:

(i) Moderate increase of carrier gives a curve which flattens out at higher values as the carrier increases.

(ii) When the increase in carrier reaches large values further increase produces little effect and the urinary excretion approaches 100 per cent in times which are short compared with those for small values of the carrier.

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NATURAL AND FALLOUT γ-ACTIVITY IN FOODSTUFFS

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Abstract—A study was undertaken to estimate the actual concentration of γ-active isotopes in vegetables, tea, tobacco, powdered milk, wheat, rice, fish, etc., which are available in Bombay, India. A large-size NaI(Tl) crystal scintillator and a thirty-channel pulse-height analyser were used to identify and to estimate the concentration of the radioisotopes. The studies were carried out over a period of about 15 months which is broadly divided as (1) the year following the stoppage of nuclear weapon tests (in October 1958), (2) the month before the French atomic weapon test in Sahara desert on February 13, 1960, and (3) the month after this test.

In addition to the presence of natural radioactivity of K^{40}, fission product activities such as Zr^{95}–Nb^{95}, Ru^{103}, I^{131}, Cs^{137}, Ba^{140}–La^{140}, Ce^{141} and Ce^{144} were noticed, but in varying amounts in different materials. The very high activities of Zr^{95}–Nb^{95} and Ce^{144} in tea were found to be chemically bound in its leaves preventing this uptake by human beings. The presence of Zr^{95}–Nb^{95} in carrots suggests its preferential pick up through the roots. The radioisotope Zn^{65} was detected in fish and prawns.

The analysis of certain vegetables during the month following the French test clearly indicated the presence of short-lived fission products which should have originated in the test. The degree of contamination is found to depend upon the total surface offered to the fallout and its roughness to hold the activity.

INTRODUCTION

Fission products from nuclear tests contaminate the biosphere and get into the human body predominantly through foodstuffs. The uptake of fission products by plants from the soil is very low owing to fixation and sorption in the soil as compared with the uptake from water solutions. This effect is very pronounced in the case of radio cesium, whereas the difference in the uptake of radiostrontium from the soil and water solution is considerably less, which is accounted for by the different degree of fixation in the soil. The fallout activities are also picked up to different degrees into the biological chain through surface contamination of leaves and other parts of the plant above the ground.[1] Fission products from the soil surface can also affect the plants through stem-base absorption.[2] The vegetation thus contaminated may be ingested directly by man, or by animals which in turn provide food for man.

The extent of incorporation of the fission products into the foodstuffs is thus rendered hard to establish, as the mode of uptake and its degree vary from plant to plant and from element to element[3,4] depending on the metabolic properties of the various elements, the type and composition of the soil on which the plants grow and the extent of the foliar and stem-base absorption, etc.

Aquatic life, on the contrary, has a remarkable property of concentrating certain elements because of their easy availability to ecological chains in aquatic as compared to terrestrial environments.[5–7] The degree of discrimination and of concentration of the elements in aquatic organism depends upon the physiological[8] demand of the organism for the elements, its availability and the chemical similarity of the elements. As the details of the mechanism of the distribution of radioisotopes in the biosphere are not adequately available, it is therefore the aim of the present study to determine the actual concentration of the various radioisotopes in the food-stuffs.

Sr^{90} and Cs^{137} are the isotopes of concern, having a high fission yield and long half-life escaping into the environment due to the

nuclear explosions, because of their biological properties such that they are readily taken up into the food chain and finally enter human beings. Sr^{90} being a β-emitter requires special techniques for quantitative analysis while γ-ray analysis of Cs^{137} is comparatively easier. The study is therefore based only on the γ-activity measurements, thereby restricting the authors to the radioisotopes like K^{40}, Zn^{65}, Zr^{95}–Nb^{95}, Ru^{103}, I^{131}, Ba^{140}–La^{140}, Ce^{141}, Ce^{144}, etc., in addition to Cs^{137}. The present report gives the results obtained for vegetables, powdered milk, wheat, rice, fish, prawn, tea, tobacco, etc., using standard methods of γ-ray spectrometry. These measurements were spread over a period of about 15 months and are divided into three different sections, viz.

(1) The year after the stoppage of nuclear weapon tests in October 1958, when fallout will mostly consist of long-lived isotopes,

(2) The month before the French atomic test of February 13, 1960, and

(3) The month after the test when contamination from short-lived isotopes can be expected. (The vegetables used for these measurements were not washed before ashing, in order to detect possible surface contamination from the fresh fallout.)

EXPERIMENTAL SET-UP

The γ-activities were measured with a low-level γ-spectrometer using a large-size NaI (Tl) crystal 4 in. dia. $\times$ 3 in. thick and a thirty-channel pulse-height analyser. The crystal was specially canned in electrolytic copper with a quartz window for low-level counting. It was optically coupled to Dumont photomultiplier type 6364 of 5 in. dia. cathode with silicone-oil type 200-DC of viscosity 100,000 centistokes. The assembly, along with the cathode follower, was shielded on all sides by 3 in. thick lead bricks. For large samples a 3-l. container, which surrounds the crystal from all sides, was used.

DETERMINATION OF THE ABSOLUTE CONCENTRATION OF THE RADIOISOTOPES

The complex γ-spectrum containing many photo-peaks was analysed by subtracting step by step, starting from the high-energy side, the contribution due to each of the photo-peaks as obtained from standard sources of similar γ-energy. In this way, the net counts in each photo-peak can be computed by eliminating the interference due to the other γ's. The absolute concentration of the particular isotopes was then determined from the relation

$$N = N_0GAER$$

where N = net counts in the photo-peak, N_0 = absolute strength of the γ-emission, G = geometry factor, A = factor to correct for the self absorption in the sample, E = detection efficiency (which has been calculated for the particular size of the crystal and for a point source with known geometry[9]) and R = ratio of the photo-peak to total counts. (This was determined experimentally for five different single-energy γ's of radioisotopes: K^{40} (1.46 MeV), Zn^{65} (1.12 MeV), Mn^{54} (840 keV), Cs^{137} (662 keV) and Be^{7} (480 keV).

The geometry factor G was determined using a standard solution of KCl having the same geometry with respect to the crystal as the samples and observing the net counts in the photo-peak of its spectrum. From the known amount of potassium in the solution which determines N_0(3.6 γ/sec per g K) and the net counts in the photo peak of its spectrum, one can determine the geometrical factor G from the above relation, which in turn can be used for other samples.

Absorption for high-energy γ's was considered negligible particularly as the maximum mass of the ashed sample was not more than 1.0 g/cm^2. In the case of low-energy γ's such as those from Ce^{141} and Ce^{144}, especially when the mass of the samples was large, suitable corrections were applied for the absorption effect.

RESULTS

The measurements reported here were made on a variety of food and other samples obtained from the local markets serving the city of Bombay. Table 1 presents measurements made on samples collected during the period January 1959 to December 1959. Table 2 gives the measurements made on vegetables in the period February 1–8, 1960 prior to the first French atomic test in Sahara. Data on the samples collected during the period March

Table 1. *Natural and fallout γ-activities in various food samples measured during January to December* 1959

Specimen	Date of acquiring	Quantity analysed (Wet wt. kg)	Amount of various radioisotopes expressed in $\mu\mu c$/kg					Potassium content (g/kg)
			Ce^{144} (285*d*)	Cs^{137} (30*y*)	Zr^{95}–Nb^{95} (65*d*–35*d*)	Zn^{65} (245*d*)	K^{40} (1.3 × 10^9*y*)	
(1) Milk powder	9 Jan.	2.70	—	65	—	—	5850	6.6
(2) Radish (roots)	12 Jan.	1.43	—	—	—	—	2740	3.1
(3) Carrots	6 Feb.	2.93	48	—	150	—	2040	2.3
(4) Tea (Nilgiri Hills)	13 Feb.	0.30	1000	470	5470	—	13,500	15.3
(5) Coffee (Nilgiri Hills)	13 Feb.	0.96	—	—	—	—	16,800	19.0
(6) Tobacco (Indian)	6 May	0.50	—	—	—	—	29,700	33.6
(7) Potatoes	19 Nov.	5.63	—	—	—	—	3540	4.0
(8) Rice	20 Nov.	1.61	—	—	—	—	1320	1.5
(9) Wheat	20 Nov.	1.60	—	—	—	—	4050	4.6
(10) Fish (Pomphret)	23 Nov.	1.72	—	—	—	65	2740	3.1
(11) Prawns	22 Dec.	2.23	—	—	—	75	2040	2.3

Table 2. *Natural and fallout γ-activities in seven different vegetables measured during February* 1–8, 1960

Specimen	Date of acquiring	Quantity analysed (Wet wt. kg)	Amount of various radioisotopes expressed in $\mu\mu c$/kg					Potassium content (g/kg)
			Ce^{144} (285*d*)	Cs^{137} (30*y*)	Zr^{95}–Nb^{95} (65*d*–35*d*)	Zn^{65} (245*d*)	K^{40} (1.3 × 10^9*y*)	
(1) Turnip	1 Feb.	4.49	—	—	—	53	1920	2.2
(2) Sugar beet	1 Feb.	5.39	—	—	—	—	3170	3.6
(3) Onions	2 Feb.	2.49	—	—	—	—	1640	1.9
(4) Yam	2 Feb.	3.48	—	—	—	—	3880	4.4
(5) Cabbage	4 Feb.	4.50	—	—	—	—	1830	2.1
(6) Cauliflower	5 Feb.	2.56	—	—	—	—	2640	3.0
(7) Spinach	8 Feb.	6.58	—	—	—	—	2650	3.0

1–16, 1960, after the first French atomic test and before the second (on April 1, 1960) are shown in Table 3. The activities recorded in the tables are the absolute concentration of the radioisotopes corrected for their modes of decay, other than the emission of γ of one particular energy.

DISCUSSION

January–December 1959

The samples collected and studied during this period were those of wheat, rice (local), carrots, potatoes, radish (roots only), powdered milk, tea, coffee, tobacco, fish and prawns.* Of these, wheat, rice, potatoes and coffee did not show any γ-activity other than that due to natural potassium (Table 1). Cs^{137} is absent

* In addition, a complete human skeleton (2.69 kg wet wt.) of a 30-yr-old male from Bombay was washed ashed and analysed for its γ-activities. It showed an activity of 450 $\mu\mu c$ of Zr^{95}–Nb^{95} per kg. This activity could have entered into the human body through certain root vegetables like carrots which show a tendency to pick up this radioisotope.

Table 3. Natural and fallout γ-activities in thirteen samples of vegetables measured during March 1–16, 1960

Specimen	Date of acquiring	Quantity analysed (Wet wt. kg)	Amount of various radioisotopes expressed in $\mu\mu$c/kg						Potassium content (g/kg)
			Ce^{141} (33*d*)	Ba^{140}–La^{140} (13*d*–40*h*)	I^{131} (8.1*d*)	Ru^{103} (40*d*)	Zr^{95}–Nb^{95} (65*d*–35*d*)	K^{40} (1.3×10^9*y*)	
(1) French beans	1 Mar.	4.56	58.4	133.6	35.4	47.6	20.4	2210	2.5
(2) Lady's fingers	2 Mar.	4.63	24.2	111.2	—	21.4	20.4	2040	2.3
(3) Brinjal	3 Mar.	4.76	7.4	—	—	17.7	—	1770	2.0
(4) Peas (seeds)	4 Mar.	2.37	—	—	—	—	—	3630	4.1
(5) Peapod	4 Mar.	1.79	56.4	364.8	—	53.8	58.5	2210	2.5
(6) Spinach	7 Mar.	4.78	120.0	480.0	103.0	149.0	100.0	2570	2.9
(7) Lettuce	10 Mar.	4.64	176.0	488.0	151.0	175.5	119.5	2390	2.7
(8) Bitter gourd (karela)	10 Mar.	4.39	21.4	—	—	14·7	—	2570	2.9
(9) White pumpkin (loki)	12 Mar.	5.26	9.9	—	—	—	—	1280	1.4
(10) Radish (leaves)	14 Mar.	1.81	79.4	128.8	79.0	87.6	76.0	3180	3.6
(11) Radish (roots)	14 Mar.	2.30	—	—	—	—	—	3890	4.4
(12) Lotus stem	15 Mar.	2.85	—	—	—	—	—	3810	4.3
(13) Marrow (torai)	16 Mar.	4.12	13.7	—	—	14.9	—	1500	1.7

in majority of the items and seems to be poorly picked up through roots by the different plants, grown in soils.[10]

Milk powder. The only fallout γ-activity found in the sample of milk powder was that of Cs which comes mostly from the surface contamination of grass ingested by cows.[9,10]

Fish and prawns. Aquatic organisms, particularly oyster, have a strong tendency to concentrate zinc.[8,11] Fish and prawns from Bombay harbour unmistakably showed the presence of Zn^{65} activity.

Tea. Samples of Indian tea were analysed and they all showed γ-activities due to Zr^{95}–Nb^{95}, Cs^{137} and Ce^{144}. It was experimentally found that the Ce^{144} and Zr^{95}–Nb^{95} activities were chemically bound* in the tea leaves, preventing their uptake by the human body. The photo-peaks of Cs^{137} and Zr^{95}–Nb^{95} were clearly separated in most of the samples when they were of moderate intensity. One particular sample whose spectrum was taken about 70 days after collection from the market showed very high activity (5470 $\mu\mu$c/kg) due to Zr^{95}–Nb^{95}. In the γ-spectrum (Fig. 1) of this sample, the photo-peak of Cs^{137} is seen merging into that of Zr^{95}–Nb^{95}. However the two peaks got separated out clearly after some time when Zr^{95}–Nb^{95} activity had decayed to some extent. The peak at about 200 keV is due to a mixture of 2 per cent γ-emission of 231 keV from Nb^{95m} (half-life 90 hr) and the back-scattered photons. The photo-peak at 490 keV is probably due to Ru^{106}.

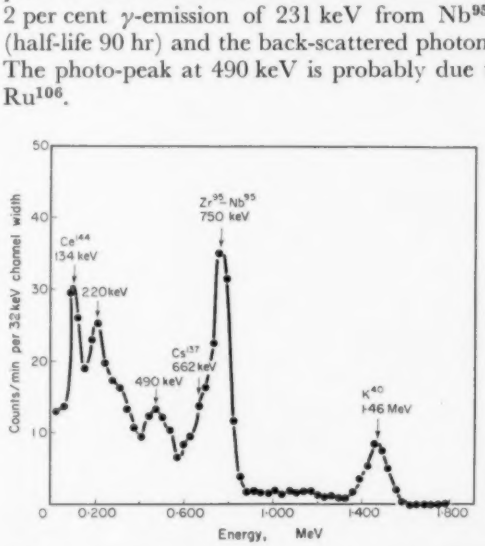

FIG. 1. γ-Spectrum of un-ashed sample (300 g) of tea leaves (Phillips) from Nilgiri Hills.

* The activities could not be washed off with water. The ashed sample of tea was brought into solution in dilute HCl, and 2% (by wt.) of NH_4Cl was added. A precipitate containing Zr^{95}–Nb^{95} and Ce^{144} activities was formed upon treatment with excess ammonia. The Cs^{137} and K^{40} activities stayed in the solution.

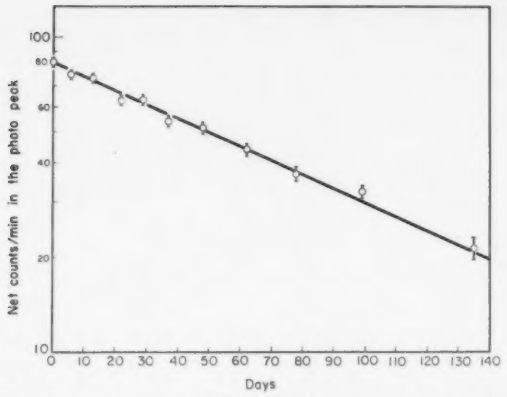

FIG. 2. Decay curve of Zr^{95}–Nb^{95} activity in a sample of tea giving a half-life of 65 days.

The Zr^{95}–Nb^{95} peak in the sample of tea was followed for about 130 days and the decay curve is plotted in Fig. 2, which gives a half-life of 65 days confirming the identity of this isotope.

The Ce^{144} and Zr^{95} activities are probably acquired as a result of absorption through surface contamination rather than due to pick-up through roots. However, it is surprising to note that tobacco, whose leaves also provide a large surface area for fallout and were collected at about the same period, did not show any detectable amount of such activities.

Carrots and radish (roots). Carrots showed an appreciable amount of Ce^{144} and Zr^{95}–Nb^{95} activities. The possible modes of uptake are:

(1) From surface contamination (which is very unlikely)[12]

(2) Absorption of the elements from the active dust accumulated at the base of the leaves

(3) Stem-base absorption

(4) Absorption through the roots

Even though radish is similar to carrots, it does not show any of these activities, thereby suggesting a preferential uptake of the isotope by carrot through the roots.

February 1–8, 1960

During this period, just prior to the French Sahara test of February 13, 1960, seven different vegetables, namely, turnip, cabbage, cauliflower, yam, spinach, sugar beet and onions were collected for analysis. None of these vegetables showed any detectable amount of fission-product activity (Table 2) within the sensitivity of the counting set-up, except for the slight indication of Zn^{65} in turnip.

March 1–16, 1960

Thirteen different samples of vegetables, namely, French beans, lady's finger, brinjal, peas (seeds), peapod, spinach, lettuce, bittergourd ("karela"), white pumpkin ("loki"), radish (leaves), radish (roots), lotus stem and marrow ("torai") were collected and assayed for their γ-activities. The results (Table 3) clearly indicated the presence of short-lived isotopes as expected. All the vegetables showed high activity due to $Zr^{95}(65d)$–$Nb^{95}(35d)$, $Ru^{103}(40d)$, $I^{131}(8.1d)$, $Ba^{140}(13d)$–$La^{140}(40H)$ and $Ce^{141}(33d)$. These are identified and confirmed by means of their γ-energies and half-lives. A γ-spectrum of lettuce is shown in Fig. 3 where the small photo-peak at 1.6 MeV due to the isotopes Ba^{140}–La^{140} is clearly seen. The

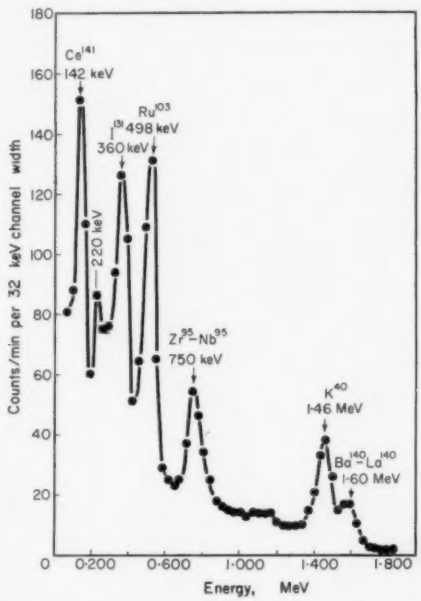

FIG. 3. γ-Spectrum of an ashed sample of lettuce taken within a month after the first French atomic test. (Shows the presence of fission products: Zr^{95}–Nb^{95}, Ru^{103}, I^{131}, Ba^{140}–La^{140}, and Ce^{141}.)

other photo-peaks due to this radioisotope at 328, 490 and 815 keV will be completely masked by the higher intensity γ's from Zr^{95}–Nb^{95}, Ru^{103} and I^{131}. The presence of these activities detected about 15–30 days after the French atomic test are in agreement with the radioisotopes expected from the fission after this much a time interval, as reported by EISENBUD,[13] confirming their origin to the above test.

The contamination levels depend upon the total surface exposed to the fallout and its roughness to hold the activities; e.g. lettuce and spinach (leafy vegetables) showed fairly high concentration of the radioisotopes as compared to other vegetables. The surface contamination of the peapod was clearly demonstrated by the absence of fission product activities in the peas (seeds) in contrast to peapod as can be seen from Table 3.

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BODY-BURDEN PROBLEM

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Abstract—If amount q of a radioactive isotope is injected in a single dose, the body burden is $R_1 = qF(t)$ where $F(t)$ is some non-negative monotone decreasing function. Similarly $R_2 = \int_0^t G(\theta)F(t-\theta)\,d\theta$ is the body burden under continuous injection rate $G(t)$. The radiation dosages D_1 and D_2 are the time integrals of R_1 and R_2. It is shown that

$$\frac{D_1}{D_2} > \frac{q}{\int_0^t G(\theta)\,d\theta} > \frac{R_1}{R_2}$$

for any $F(t)$ with the above-stated properties and any non-negative integrable $G(t)$. The implications of these inequalities are discussed.

In a discussion of retention of radioactive bone seekers with special reference to the utility of the power function as a formal description of retention or body burden, Norris *et al.*[1] note that use of the power function description leads to major disparities in radiation dosage depending upon the mode of administration (i.e. condition of exposure or manner in which a given body burden is attained). In particular they show that at the time at which equal body burdens occur with (i) a single administration and (ii) a continuous administration at a constant rate, the radiation dosage in the first case exceeds that in the second. This conclusion was reached by direct integration of the appropriate empirical function, and the quantitative disparity between the radiation dosages depends upon an estimated parameter of the power function.

It is the purpose of this paper to point out that the qualitative conclusion can be reached under much more general circumstances, viz., when we consider any acceptable (a term made precise in what follows) "unloading function", and compare a single administration with continuous administration at any integrable, but otherwise arbitrary, rate. Moreover, this conclusion is a special case of a more general set of inequalities relating the body burdens at any time, the integrated radiation dosages at that time and the amounts of material administered up to that time.

If a single administration of amount q is given at time zero, the retention or body burden is given by

$$R_1(t) = qF(t) \tag{1}$$

while if the radioactive material is given at a rate $G(t)$, the retention is

$$R_2(t) = G(t) * F(t) \tag{2}$$

where for any two functions $M(t)$, $N(t)$ the convolution is denoted in the usual way by

$$N * M = M * N = \int_0^t M(\theta)N(t-\theta)\,d\theta \tag{3}$$

In (1) and (2), $F(t)$, the unloading function, has physical significance of the fraction of an amount administered at time zero which remains at any subsequent time t and must possess the properties $1 \geqq F(t) \geqq 0$, $dF/dt \leqq 0$, (in fact $F(0) = 1$, $F(\infty) = 0$, but we make use only of the non-negative and monotone decreasing properties), and $G(t)$ is a non-negative integrable function. It must further be assumed that $F(t)$

is independent of q and that the system possesses the requisite properties of linearity which ensure the validity of the convolution (2) (see Refs. 2–4 for discussion).† We note in passing that under the hypothesis of a compartmental model

$$F(t) = \Sigma a_i e^{-\lambda_i t},\ \lambda_i > 0, \quad \Sigma a_i = 1$$

and each of the properties and assumptions mentioned above is satisfied.

In order to discuss a situation comparable to that considered by NORRIS *et al.*[1], we eschew questions of lack of uniformity of distribution of material and irradiation, and assert the integrated radiation dosage, hereafter referred to simply as dose, to be proportional§ to

$$D_1(t) = q * F(t) \tag{4}$$

corresponding to (1), and to

$$D_2(t) = 1 * G(t) * F(t) \tag{5}$$

corresponding to (2). We note from (3) that if N is a non-negative function, then

$$M * N < M(1 * N) \tag{6}$$

if M is an increasing function, while

$$M * N > M(1 * N) \tag{7}$$

if M is a decreasing function.‡ Applying (7) to (2), on the basis that G is non-negative and F is decreasing, there follows

$$R_2 > F(1 * G) = R_1(1 * G)/q \tag{8}$$

and applying (6) to (5), on the basis that F is non-negative and $G \geqq 0$ requires $1 * G$ to be increasing, there follows

$$D_2 < (1 * F)(1 * G) = D_1(1 * G)/q \tag{9}$$

From (8) and (9), we have for any $t > 0$,

$$\frac{D_1(t)}{D_2(t)} > \frac{q}{Q(t)} > \frac{R_1(t)}{R_2(t)} \tag{10}$$

where $Q(t) = 1 * G$ is the quantity of material administered up to time t under the continuous injection rate $G(t)$.

The crux of the relation between R_1 and R_2 and between D_1 and D_2 is found in the fact that R_2 is obtainable from R_1 (a decreasing function) and D_2 is obtainable from D_1 (an increasing function) by the same operation, viz., convolution with the non-negative function $G(t)/q$:

$$R_1 * G/q = R_2, \quad D_1 * G/q = D_2$$

Application of (7) and (6) to these relations respectively yields, of course, the inequalities (10).

From (10) it is seen that $R_1 = R_2$ implies $D_1 > D_2$ and this is a generalization of the NORRIS–TYLER–BRUES result which they obtain specifically from $D_1/D_2 = q(b + 2)/Q$, with the condition $q/Q = 1/(b + 1)$ when equal body burdens obtain (here $b < 0$ is the exponent in the power function, $Q = Ct$ with C the constant injection rate and the decay correction has been neglected). In general $R_1 \geqq R_2$ implies both $D_1 > D_2$ and $q > Q$; $D_2 \geqq D_1$ implies $R_2 > R_1$ and $Q > q$; $q \geqq Q$ implies $D_1 > D_2$ but nothing about R_1/R_2; while $Q \geqq q$ requires $R_2 > R_1$ but tells nothing of D_1/D_2. Equal amounts administered, $q = Q$, requires both $D_1 > D_2$ and $R_2 > R_1$.

If $G(t) = C$, a constant, then at that time at which $R_1 = R_2$, the ratio of doses is given by $D_1/D_2 = (1 * F)^2/F(t * F)$, a quantity which we have proved to exceed unity for any monotone decreasing F, and this is the most general form of the case considered by NORRIS *et al.*

It is perhaps of interest to note that if a compartmental model is adopted then the amount in the kth compartment is $qF_k(t)$, for a single administration of amount q. Except for that k corresponding to the compartment receiving

† We are assuming that the decay factor $f(t) = \exp(-\lambda t)$, where λ is the appropriate decay constant, is included as a multiplicative factor. This assumption changes none of the stated properties of $F(t)$. Further, this assumption is correct when the conditions ensuring the validity of (2) are met and not otherwise.

§ Since we will deal only with the ratio of doses or inequalities between doses, any such statement regarding D_1 and D_2 permits an equivalent statement regarding doses.

‡ In addition to (6) and (7) we have $M * N > M * P$ if $N > P$, $M \geqq 0$; $M * (PN) > P(M * N)$ if M, $N \geqq 0$, P decreasing. Application of these, is an obvious sequence, to $1 * F * G$ yields $(1 * F)(1 * G) > 1 * F * G > 1 * [F(1 * G)] > F(t * G)$ and while we employ only the first inequality in this chain the remaining ones may be of use in comparing doses and body burdens under two different injection rates say G_1 and G_2.

the administration, $F_k(t)$ is *not* monotone decreasing, although $F(t) = \Sigma F_k(t)$ does of necessity possess that property.§ If we consider the dose to a given site or compartment the inequality corresponding to (9) is still obtained and hence the *first* inequality of (10) holds for any compartment. For (9) depends upon $F \geqq 0$ and not upon monotonicity. Thus $q \geqq Q$ implies $D_1 > D_2$ for any compartment. At that time at which equal amounts of material have been administered to the whole body the radiation dosage in any compartment is greater under single injection than under continuous administration. The relation between the *compartmental* burdens, dosages, and amounts of material administered (corresponding to (10) for the whole-body case) is a matter for further study.

§ Strictly speaking there is a square array of functions $F_{kj}(t)$ giving the amount at time t in compartment k when unit amount is initially present in compartment j. For $k \neq j$, $F_{kj}(t)$ is usually unimodal and $F_{kk}(t)$ is usually monotone decreasing. Conditions under which F_{kj} is approximately independent of j will be the subject of a future note.

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A COMPARISON OF HTO IN PLASMA AND EXPIRED WATER VAPOR

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Abstract—Tritium-labeled water was injected intravenously into a young male adult and the time courses of specific radioactivity in plasma, red cells, urine and expired water vapor were determined and compared. Specific activity of breath water and urine appeared to follow a pattern of damped oscillation about the curve for the specific activity of plasma water. The transfer function of the biological system for the injection of radioactivity was determined. Results indicated that breath water samples may under certain circumstances be substituted for urine or plasma water samples in the determination of total body water.

INTRODUCTION

THE dilution volume of tritium-labeled water (HTO) is often used to estimate the volume of total exchangeable body water. The magnitude of the tritium space is usually obtained by extrapolation from HTO specific activity in serial plasma[1] or urine samples[2] following injection of a known amount of HTO. In certain instances, these sampling procedures may be either difficult to accomplish or too time-consuming. With obese subjects, for example, obtaining blood samples at planned intervals may be impossible, particularly if venepunctures are routinely necessary for other analytical procedures. Accessible veins sometimes become sclerosed after frequent sampling, and still further venepuncture may be impracticable. Bladder catheterization to insure complete timed collections of urine is not an innocuous procedure. Voluntary voiding at fixed intervals to insure an adequate urine volume at each specified time has proved to be a reasonably satisfactory sampling method, but prior water loading is necessary. While water loading is easy to accomplish, it nevertheless requires preparation of measured volumes of water and extends the procedure by the time required for the first water administration. In view of these problems and in consideration of PINSON and LANGHAM'S[2] conclusion that HTO vapor in expired air is in equilibrium with HTO concentration in plasma, it was decided to explore the feasibility and practicality of collection of HTO–H_2O from expired air as a simple sampling procedure for the determination of total body water. Following intravenous administration of HTO, the time course of HTO concentration in plasma, urine and breath water of the subject was followed. Analytic expressions were found to express the relation between concentration and time for the fluids studied.

APPARATUS

The apparatus consisted of an inspired and an expired air-vapor trap plus a mouthpiece and valve system to insure unilateral air flow. The vapor traps were placed in separate dewar flasks and were cooled in a dry ice and ethanol bath. (Traps were vacuum vapor traps, catalog No. JV-8650, Scientific Glass Apparatus Co., Bloomfield, N. J.) The first trap dried the air inhaled by the subject, and the second trap condensed and collected the water vapor he expired. Ten min of breathing through the system produced a sufficiently large volume (1.5 ml) of HTO–H_2O for subsequent analysis.

* This work was done while the author was on the staff of the National Institute of Arthritis and Metabolic Diseases, National Institutes of Health, Bethesda, Maryland.

METHODS AND MATERIALS

Two complete experiments were performed on a single subject. The subject was a healthy young male, age 21, who weighed 72.7 kg. On the morning of a test day, water intake of the subject was controlled for 2 hr prior to the tracer injection and he was allowed nothing to eat following the meal given at 5–6 p.m. the previous evening. Each hour after the injection, 150 ml of tap water were given to insure adequate urine collection. The subject was confined to his bed except to void or to permit sampling.

When control samples of blood, urine and breath water had been collected, about 640 mc of undiluted HTO (gravimetrically measured dose of pyrogen-free tritiated water, lot T-003-1, Abbott Laboratories, Inc., North Chicago, Ill.) in a volume of about 1.4 ml was injected into an antecubital vein. Prior to withdrawing the needle from the vein, the syringe was flushed five times with blood. Subsequent blood samples were removed from the contralateral antecubital vein. Blood was transferred to a centrifuge tube containing a few crystals of heparin and centrifuged. One ml of plasma was transferred to a 50-ml Florence flask and shell-frozen around the interior of the flask. After centrifugation, the red cells were shaken vigorously with dry glass beads, and 1 ml of the fluid resulting was saved for further treatment. Plasma, red cell water and urine samples were shell-frozen around the inner surface of 50-ml Florence flasks. These flasks were then attached to a vacuum sublimation system[1] in which the sublimates were collected in vapor traps at −74°C for subsequent radioassay.

A liquid scintillation spectrometer was used for the radioassay method. The phosphor consisted of 6 g diphenyloxazole (DPO), 0.2 g of POPOP, and 250 ml of absolute ethanol per liter of toluene. 0.1 ml of the aqueous sample was added to 15 ml of phosphor, mixed, stored in the dark at 12°F for at least 24 hr prior to radioassay to permit decay of ultraviolet-induced phosphorescence. The efficiency of counting was about 11 per cent.[1] Standards were made by diluting an aliquot from the same lot of tritiated water used for the injection. The radiation measured from vials containing the standard aliquots decreased with time slightly faster than expected from radioactive decay alone. Because of this "aging" effect, new standard and background samples were made for each set of radioassays.

RESULTS

The results of experiment 1 appear in Table 1 and Figs. 1 and 2, which show the time course of tritium concentration in breath water and plasma. The specific radioactivity of plasma water decreased monotonically except for a deviation which occurred 30 min after injection. The highest concentration of radioactivity found in breath water was that in a sample taken 9 min

Table 1. *Specific radioactivity in body fluids* (*Subject R.A.*; *Experiment* 1; 634 *μc in* 1.3766 *g of water was injected at* $t = 0$)

Time elapsed since injection (min)	Net specific activity in Breath water (mμc/g)	Plasma (mμc/g)	Sp. Act. B.W./Sp. Act. Plasma
0	0.27	0.5	0.54
5	24.0	27.6	0.87
11	25.3	20.7	1.22
21	17.6	16.0	1.10
30	15.7	16.1	0.97
45	14.6	15.4	0.95
61	14.0	14.7	0.95
174	13.7	14.6	0.94
294	0	13.9	—
81,000	0	0.48	—

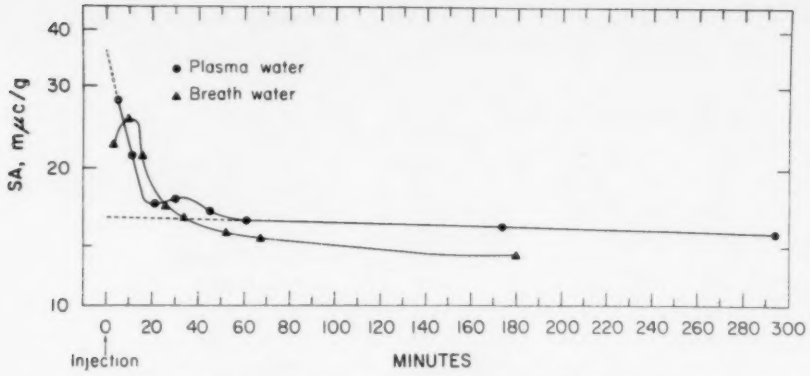

FIG. 1. Specific radioactivity of body fluid vs. time. Subject: R.A.; Expt. No. 1; Feb. 5, 1960. The subject received an injection of 634 μc of tritium-labeled water at time $t = 0$. Serial samples were taken of plasma and expired water vapor. Water was separated from the samples of plasma by low temperature, low pressure sublimation. The samples were radioassayed. Specific activity (SA) in mμc/g of water was calculated for the samples of plasma water (○) and breath water (△).

after injection. Specific activity of subsequent samples decreased regularly.

French curves were used to draw a smooth line interconnecting the data points. From the curve so drawn for plasma water (points identified by filled circles), three exponential terms (not shown) were resolved with half-lives of 0.07, 1 and 41 hr, respectively.

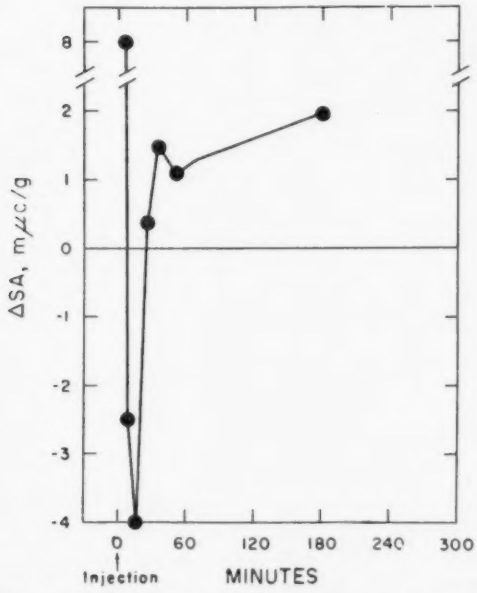

FIG. 2. Difference between specific radioactivity (△SA) in plasma and breath water vs. time. Subject: R.A.; Expt. No. 1; Feb. 5, 1960.

Fig. 2 shows the differences between specific radioactivities in plasma and breath water for this experiment. The specific activity of breath water was at first less, then greater, and finally less than the corresponding specific activity of plasma water sampled at the same time. During the initial part of the experiment, there was a delay of about 6 min between the time when the plasma specific activity was a given value and the time when the specific activity of the breath was the same value. The frequency of sampling was too low to detect high-frequency components in the transient response, but the curve crossed the line of zero concentration difference at least twice.

The results of experiment 2 are shown in Table 2 and in Figs. 3 and 4. Fig. 3 shows the results from a repetition of experiment 1 under the same conditions on the same subject. A deviation from the monotonic decrease in plasma radioactivity occurred at about 32 min after injection, which resembled the deviation in the first experiment in both amplitude and time of occurrence.

Table 2. Specific radioactivity in body fluids (Subject R.A.; Experiment 2; 644 μc in 1.4119 g of water was injected at t = 0)

Time elapsed since injection (min)	Net specific activity in Breath water (mμc/g)	Plasma (mμc/g)	Sp. Act. B.W./Sp. Act. Plasma
0	0.7	0.7	1.00
7	27.4	22.3	1.23
13	24.3	18.9	1.29
18	20.3	16.4	1.23
25	17.5	15.7	1.12
31	16.0	15.8	1.01
37	15.2	15.7	0.97
48	14.6	14.4	1.01
59	14.1	14.4	0.98
79	13.5	14.5	0.93
134	12.3	14.0	0.88
195	12.2	12.9	0.95
252	12.1	12.9	0.94
320	—	12.6	—
62,000	—	0.68	—

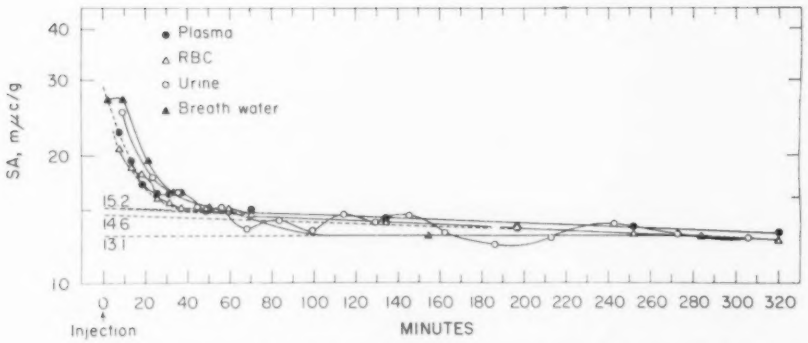

FIG. 3. Specific radioactivity of body fluids vs. time. Subject: R.A.; Expt. No. 2; March 17, 1960. The subject received an injection of 644 μc of tritium-labeled water at time t = 0. Serial samples were taken of blood, urine and expired water vapor. Water was separated from the samples of plasma, crushed red cells and urine by low temperature, low pressure sublimation. The samples were radioassayed. Specific activity (SA), in mμc/g of water was calculated for the samples of plasma water, (●), red cell water (△), urine (○) and expired breath water (▲).

The highest concentration of radioactivity found in breath water was that in the sample taken 10 min after injection. During the early part of this experiment, there was delay of 6–8 min between the time when the concentration in plasma was a given value to the time when the concentration in breath water was the same value. French curves were used to draw a smooth line through the experimentally obtained points. From a smooth line drawn for plasma water (points identified by filled circles), a series of three exponential terms (not shown in Fig. 4) were resolved. These terms had half-lives of 0.08, 2 and 27 hr, respectively.

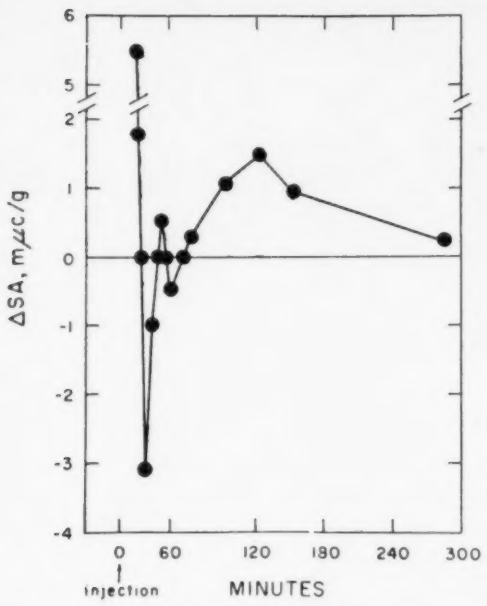

FIG. 4. Difference between specific radioactivity (△SA) in plasma and breath water vs. time. Subject: R.A.; Expt. No. 2; March 17, 1960.

In Fig. 3, the specific activities of the water extracted from urine and from red cells were also plotted. After the initial mixing periods, the curve of urine specific activity oscillated about the curve of plasma specific activity. (This may reflect either the hourly ingestion of 150 ml of water, the frequency of voiding, intradiurnal variation of urine elaboration, or some combination of these rhythms.) The curve of activity in red cell water shows the specific activity in one type of cell. This curve follows closely the curve for plasma water.

Table 3 gives estimates of tritium space based on radioassays of samples from various body fluids. Estimates based on breath water were regularly about 10 per cent higher than estimates based on plasma (taken to be the reference fluid). The body water volume estimates from breath water results were multiplied by 0.94 to yield a "corrected" estimate of tritium space. The tritium space of the subject was 60–61 per cent of total body weight. This is consistent with the 62.2 per cent reported by PINSON and LANGHAM[2] and the 61.8 per cent reported by SCHLOERB *et al.*[3]

Fig. 4 shows the difference between specific radioactivities in plasma and breath water for this experiment. A comparison of Tables 1 and 2 shows that the frequency of sampling was higher in experiment 2 than in experiment 1, and better resolution of cycles of transient response was obtained. The difference curve crossed the time axis at least four times before being damped and approaching a steady-state value.

The author knows of no laboratory which uses a smaller dose of tritium to estimate adult human body water volume. The work reported here demonstrated that a dose this low (0.6 mc) gave data which are quantitatively comparable to those obtained by the administration of tracer doses two or more times larger. By using a minimal amount of radioactivity, unnecessary radiation exposure of the subject is avoided.* This is particularly desirable if repeated determinations are to be performed on the same subject. A dose of 634 mc produced a whole body radiation dose of 52 mrems for the first week and 157 mrems for a total dose.

DISCUSSION

Fig. 2 shows the differences between the specific activities of plasma and of breath water versus time for experiment 1. Fig. 4 shows the corresponding differences versus time for experiment 2. Insofar as these different curves are significant, they resemble a damped oscillation. SHREEVE *et al.*[5] have detected such oscillations in a similar experiment using $C^{14}O_2$. In terms of this compartmental model, oscillations could be due to non-linear transfer coefficients between compartments.

* The whole body tritium radiation dose was calculated[4] to be

$$D = 88ET_{eff}C(1 - \epsilon^{-\lambda_{eff}t}) \times \text{RBE}$$

where D = dose (rems), E = average energy of beta particles (MeV = 0.006), T_{eff} = effective half-life (days) = 12, C = concentration of radioisotope in critical organ (μc/g) = 634/(43,400 × 0.9933) = 0.0147, λ_{eff} = effective decay constant (days^{-1}) = 0.0578, t = exposure time (days), RBE = relative biological effectiveness = (1.7),[2] D = 52 mrems the first week (this is less than half the allowable dose for radiation workers: 5000 mrems/year/50 weeks = 100 mrems/week) = 157 mrems for the total dose.

Table 3. Comparison of body water volumes as estimated from sampling various body fluids

Experiment	Plasma (l.)	Breath waters (l.)	"Corrected" breath waters (l.)	Urine waters (l.)	Red cell waters (l.)	Part of BWVs (%)
1	43.57	48.18	45.3	—	—	60
2	44.1	50.1	47.1	52	50.0	61

The body water volume estimates from breath water data were multiplied by 0.94 to yield "corrected"estimates of tritium space.

The interpretation of the specific radioactivity of the breath water is complicated by several factors. Expired air is a mixture of the air of the alveoli and of the dead space, and water vapor is added and exchanged along the entire respiratory tract and in the lungs. Therefore, the specific activity of expired water vapor cannot be uniquely assigned to a given anatomical site or time after injection; the sample includes concentrations valid for a range of times.

In each experiment, the curve for breath water concentration crossed the curve for plasma water concentration about 30 min post-injection. After the initial period, the values converged gradually. One reason for the lower specific activity in breath water may have been the decreased diffusion mobility of the heavier tagged water molecule compared with untagged water. According to Graham's law, the rate of diffusion of a labeled water molecule is about 5 per cent less than that of an unlabeled molecule. The results show that during the period from 1 to 5 hr after injection, the specific activity in breath water was 88–98 per cent of the specific activity in plasma.

Pinson and Langham[2] reported that when a subject was exposed to gaseous tritium, it was readily observed in the blood. In the present paper, the reverse movement was studied: movement of tritium from the blood to the atmosphere. The results reported here indicate that the concentration of tritium in expired breath was 5–10 per cent lower than the concentration of tritium in blood. If this were due to a mechanism which was independent of direction, then one might expect that if a subject were exposed to an atmosphere containing tritium water vapor, after an hour the concentration in the blood leaving the lungs would be 5–10 per cent lower than the concentration in the inspired breath.

A corresponding isotope effect was detected *in vitro*. When tritium-labeled water was fractionally sublimated *in vitro*, the difference in HTO concentration between the first and the last sample was about 6 per cent. This suggests that the migration velocity of the labeled molecules was sufficiently different from that of the unlabeled molecules to cause a significant and detectable difference in the results, depending on when samples were obtained.

The interpretation of urine specific activity is not clear. Water in the plasma moved across Bowman's capsule to form the bulk of the urine volume. However, in the tubule, water was resorbed, and the amount of resorption was strongly dependent on the concentration of electrolytes and certain hormones in the blood. Mixing occurred with previously elaborated urine in the bladder, and there was a variable time delay before voiding. Also, the specific activity of urine appeared to oscillate with each bout of drinking by the subject.

Breath water vapor collection offers a decided advantage as a sample collecting method because: venepuncture is avoided, and there is no need to remove proteins or coloring substances from the sample because the patient had performed a purifying evaporation for the investigator, and samples can be collected as frequently as necessary (limited only by time required to collect sufficient expired water vapor for analysis, 5–10 min). Consequently, analysis results were available sooner and cheaper. Breath water samples may be collected more frequently than urine samples unless catherization is used.

Table 3 shows the estimates of body water

volume obtained from sampling these body fluids. In this laboratory, breath water samples gave more reliable and reproducible results than urine water samples, even after subliming the water from the urine to radioassay the pure water.

PINSON and LANGHAM's paper[2] mentions a 90-day half-life for tritium atoms reincorporated into protein molecules. If this is accepted, it would contribute a fifth exponential term with an elimination constant of -0.00032 in the units of the other elimination constant, hr^{-1}. Thus, for experiment 1, the graph showing the concentration of tritium in plasma water vs. time was resolved into three exponential terms. Data from samples taken a few weeks after injection were analyzed to give a fourth exponential term. The 90-day half-life for tritium atoms reincorporated into protein molecules gave a fifth exponential term. For experiment 1, the time course of tritium concentration is given by the sum of these five terms, which is

$$Cp = 18.6 \exp(-10.3t) + 1.31 \exp(-0.68t) + 2.4 \exp(-0.017t) + 13.2 \exp(-0.0025t) + 0.6 \exp(-0.00032t)$$

where Cp = the concentration of tritium in plasma water (mμc per g) and t = the time elapsed since injection (hr).

The corresponding equation for experiment 2 is:

$$Cp = 12.6 \exp(-8.6t) + 2.2 \exp(-0.33t) + 0.9 \exp(-0.025t) + 13.3 \exp(-0.0025t) + 0.6 \exp(-0.00032t)$$

Fig. 5 shows how well the values for specific activity calculated from the postulated model and above formula agree with the experimental data.

The experimental curves deviate from a smooth sum of exponentials at a small peak occurring about 30 min after injection. One may view the subject as a multi-compartment biological system subject to a small perturbation by the injection of the tracer. The transfer function of a system is an expression relating output of a signal (in this instance, the radioactive tracer) to the input. This experiment is an interesting example of how one may view the subject as a control system and obtain a fundamental parameter of the system by the observation of its performance. For this experiment,

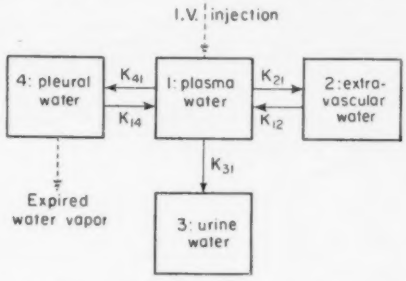

FIG. 5. Comparison of calculated and experimental values for specific activities of lung water and plasma water for Expt. 2.

the transfer function describes the response of the system, and it is given simply by the above equations for the two experiments, respectively.[6]

Fig. 6 shows a postulated model for the handling of water by this subject. The data were used to estimate compartment sizes and transfer coefficients using an iterative program by BERMAN[10] and the IBM 704 computer of the National Bureau of Standards. Approximate values found were:

K_1, plasma HTO space + extracellular HTO space = 44 ± 2 l., or 60% of body weight

K_2, lung HTO space = 0.26 l., or 0.36% of body weight

K_3, urinary HTO space = open

K_4, intracellular HTO space = 12 l., or 16.6% of body weight

The values of the transfer coefficients were:

$$k_{12} = 0.043 \pm 0.006 \text{ min}^{-1}$$
$$k_{21} = 0.043 \pm 0.009$$
$$k_{31} = 0.0013 \pm 0.0003$$
$$k_{14} = 0.770$$
$$k_{41} = 0.0048 \pm 0.0002$$

Using this model, the values of plasma water specific activity were calculated for times corresponding to the times at which blood samples

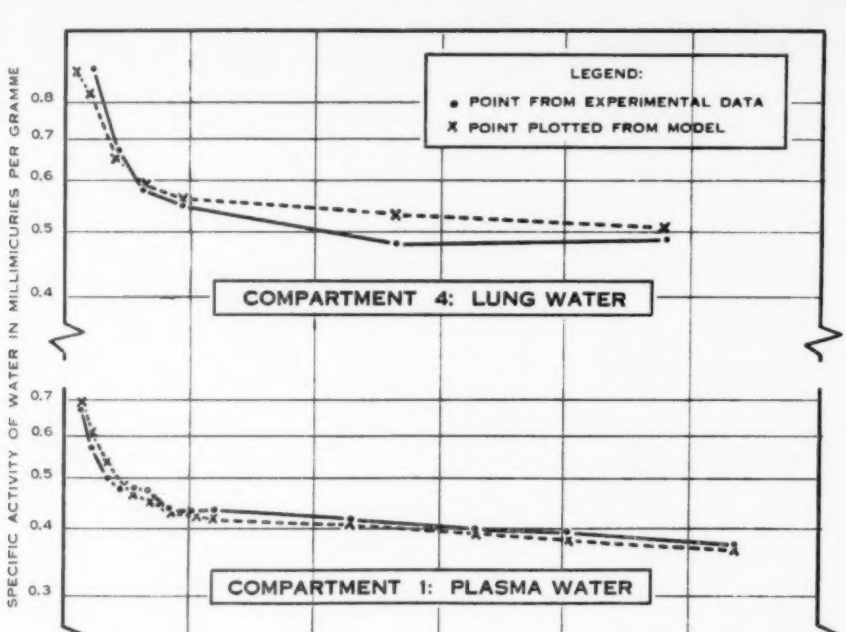

FIG. 6. Postulated four-compartment model for water distribution in man. (k_{ij} in the coefficient of transfer of water to compartment i from compartment j).

had been drawn. The comparison of the calculated and experimental values is shown in Fig. 5.

SUMMARY

(1) Three hr or longer after injection, the concentration of tritium in expired water vapor was about 94 per cent of that in the blood plasma, while the concentration in urine equaled that in plasma under the conditions of this study.

(2) The concentration of tritium in the water of red cells was consistently a little less than that in plasma when the two were measured simultaneously at any time after injection.

(3) The plasma concentration curve can be described as the sum of five exponentials with decay constants corresponding to half-lives of the order of $\frac{1}{3}$ hr, 3.6 hr, 46 hr, 12 days and 90 days, respectively.

(4) A departure of the curve from this exponential decay occurred at $\frac{1}{2}$ hr after injection, suggesting an unknown and complicated movement of tracer at this time.

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MEASUREMENT OF BONE-MARROW DOSE IN A HUMAN PHANTOM FOR Co^{60} γ-RAYS AND LOW-ENERGY X-RAYS*

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INTRODUCTION

FOR estimating the biological effects that would result from doses of X-rays and γ-rays of differing photon spectra, and for comparing experimental data obtained using different irradiation sources, various concepts of specifying the effective dose have been used. One of the more widely adopted has been the integral body dose postulated by MAYNEORD[1]. The exit dose[2,3] and the midline tissue dose[4,5] have also proved useful.

In view of the generally accepted belief that the dose to the hematopoietic organs is critical from the point of view of acute lethality, it is considered that a better criterion would be bone-marrow tissue dose. However, there is little experimental data on the absorbed dose in bone marrow, and most of what is available is concerned with discrete experimental conditions simulating diagnostic or therapeutic procedures. It was therefore considered desirable to obtain additional data on bone-marrow dose for various photon energies, using a standardized irradiation procedure.

In measuring the absorbed dose in bone-enclosed tissue there are two opposing effects to be considered, the shielding of the marrow by the bone structure, and the enhancement of dose in the marrow which is close to the bone structure. However, enhancement is discussed only briefly because the effect is expected to result in an average increase in marrow dose of only a few per cent in the range of photon energies used. The decrease in marrow dose due to shielding by the bone is therefore the main effect considered.

The experiment reported here is designed to give information on the bone-marrow dose obtained from a uniformly distributed source of radiation, such as one would obtain from fallout. Human bone was contained within the phantom to be irradiated, and seven different exposure conditions were used including γ-rays from Co^{60} and X-rays generated at potentials of 50–250 kV. The field size was greater than the phantom dimensions, and the phantom was rotated continuously during irradiation at a distance of about 1.5 m from the source.

The requirements on the dosimeter for this type of measurement are particularly stringent. The sensitive chemical system recently reported by ARMSTRONG and GRANT[6,7] has permitted obtaining the data in a reasonable time, and the energy dependence over the photon-energy range of interest is probably less than ± 15 per cent. The volume of the dosimeter was kept as small as possible so that the field in the bone would be disturbed very little by the presence of the dosimeter.

THE DOSIMETER SYSTEM

1. *Technique*

The basic chemical system, developed by ARMSTRONG and GRANT[6,7] is a 10^{-4} molar solution of calcium benzoate in water. On irradiation, salicylic acid is produced and there is a linear relationship between absorbed dose and salicylic acid concentration. The analysis for salicylic acid concentration is carried out on

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an Aminco Bowman spectrophotofluorometer* at an activating wavelength of 290 mμ and an activated wavelength of 400 mμ. Calcium benzoate fluoresces but slightly at these wavelengths. In this work, small volume cells (0.1 ml) were employed. Fig. 1 illustrates the linear relationship between salicylic acid concentration in irradiated calcium benzoate solutions and the absorbed dose in rads in the 0–400 rad region for the small-volume dosimeter.†

The irradiation cells were of Pyrex glass of length 4 cm, i.d. 2.2 $\pm$ 0.2 mm and wall thickness 0.20 $\pm$ 0.01 mm (Fig. 2). All solutions were prepared from twice-distilled water, the final distillation being from alkaline permanganate solution. The calcium benzoate was obtained from the interaction of reagent grade calcium chloride and sodium benzoate. Purification by recrystallization from twice-distilled water was carried out several times. All glassware was cleaned using fresh chromic acid followed by rinsing with twice-distilled water and steaming. In addition, the irradiation cells were heated in an oxygen flame. For irradiation of short durations the cell end was sealed with cellulose tape.

2. *Energy dependence*

The energy dependence of the salicylic acid dosimeter was determined by direct comparison with the ferrous sulphate system at the photon energies employed. Below 100 kVp, 0.1 N sulphuric acid was substituted for the 0.8 N sulphuric acid to minimize photo-electric absorption by sulphur. In order to compare the various experimental values reported, the following technique was employed. The photon distribution of X-ray spectra from 100 kVp with slight filtration to 260 kVp with medium filtration was calculated from Kramers' theoretical distribution

$$I(E)\,dE = C(E_0 - E)\,dE \prod_i \times \exp - \left\{ \left[\frac{\mu}{\rho}(E) \right]_i \rho_i x_i \right\}$$

where E_0 is the peak energy, $\left[\frac{\mu}{\rho}(E)\right]_i$ is the mass absorption coefficient of the ith filter of density ρ_i and thickness x_i. For spectra from 50 to 100 kVp experimental distribution curves of AITKEN and DIXON were used.[8] Then, using the data of JOHNS *et al.*,[9] the initial energy spectrum of electrons set in motion in water by the various X-ray spectra was obtained. From this distribution the mean linear energy transfer of the initial electron spectrum was calculated using the Bethe–Bloch formula for electron-energy deposition. Table 1 gives the mean LET of typical spectra. Col. 3 is a G-value calculated from an empirical relationship established by SCHULER and ALLEN[10] for deuterons. It is assumed that the relationship holds for electrons of the same LET.

Table 1. Mean linear energy transfer in water of various X-ray spectra

Spectrum	Mean LET (eV/A°)	Calc. G_{Fe} (ions/ 100 eV)
Tritium β's	0.55	12.7
50 kVp 1 mm Al filtration	0.65	12.7
60 kVp 1.5 mm Al filtration	0.67	12.6
80 kVp 2 mm Al filtration	0.62	12.6
100 kVp 2.5 mm Al filtration	0.60	12.7
160 kVp 0.5 mm Cu filtration	0.40	13.3
200 kVp 1.0 mm Cu filtration	0.34	13.6
260 kVp 1.0 mm Cu filtration	0.30	13.9
Co^{60} γ's	2.7×10^{-2}	15.5

Table 2 lists experimental ferrous sulphate G-values[11–19] with an assigned mean LET and a calculated G-value.

It is interesting to note from Table 1 that, despite the widely differing X-ray spectra, the mean LET of the initial electron spectra differ but slightly, especially below 100 kVp. It would be expected that on this account experimental G-values in this region would not differ greatly, but available results for ferrous sulphate do in fact vary from 12.3 ions/100eV to 14.7 ions/100 eV.

On the basis of this analysis, ferrous sulphate G-values have been assigned (Table 3). The values of G_{sal}, for the experimental spectra used,

* The slit program used was No. 4 with the modifications that slit No. 3 was replaced by a $\frac{1}{16}$-in. slit and slit No. 4 by a $\frac{3}{32}$-in. slit. This modification was found to give the maximum sensitivity whilst maintaining a low calcium benzoate blank reading.

† Figs. 1–4 were not received until this article was in press and are therefore published at the end of the article.

Table 2. Experimental and calculated ferrous sulphate G-values for ionizing radiation of different LET

Ref.	Spectrum	LET (eV/A°)	Exp. *G*-values	Calc. *G*-values
11	$H_3\beta$	0.55	12.9	12.6
12	$H_3\beta$	0.55	12.7	12.6
13	60 kVp unfilt.	0.65	13.1	12.7
14	50 kVp unfilt.	0.65	14.0	12.7
13	100 kVp, 0.65 mm Al	0.60	14.7	12.7
15	300 kVp	0.30	14.8	13.9
	100 kVp	0.60	12.3	12.7
16	1.25 MeV	2.7×10^{-2}	15.6	15.5
17	50 kVp, 0.25 mm Al	0.65	14.4	12.7
18	200 kVp (100 keV eff.)	0.34	15.3	13.6
13	220 kVp, 0.65 mm Al	0.34	15.0	13.6
19	200 kVp (75 keV eff.)	0.34	14.4	13.6

determined by comparison with the ferrous–ferric dosimeter, are given in Table 4. Reproducibility of the ferrous–ferric dosimeter system has been found to be about ± 2 per cent while that of the measurement of salicylic acid concentration is about ± 3 per cent. The estimated total uncertainty in the values of G_{sal} determined by comparison with the ferrous–ferric dosimeter is therefore not greater than ± 10 per cent at low photon energies decreasing to ± 5 per cent at high energies.

Table 3. Values of G_{Fe} assigned at different photon energies

Energy	Assigned G_{Fe} (ions/100 eV)
1.25 MeV (Co^{60})	15.6 ± 0.1
200–300 kVp X-ray spectra	14.9 ± 0.5
100–200 kVp X-ray spectra	14.2 ± 0.7
50–100 kVp X-ray spectra	13.5 ± 0.7

Table 4. G-Value vs. photon energy for the salicylic acid dosimeter

Photon spectrum	G_{sal}
Co^{60}	0.64
250 kVp	0.57
150 kVp	0.52
100 kVp	0.52
80 kVp	0.52
60 kVp	0.52
50 kVp	0.52

When the dosimeter is used to measure the dose in the phantom bones the spectral distribution at the dosimeter site is modified from that of the incident radiation. Backscatter in the phantom tends to degrade the radiation, while attenuation in the bone tends to harden the mixed spectrum which is incident on the bone site. If the energy dependence of the dosimeter were negligible, no error would be attributable to the altered spectrum, but the decrease with photon energy of G_{sal} from 0.64 to 0.52 means that some error may be present in the measurements due to this cause.

However, from the values for G_{sal} quoted in Table 4 it would appear that for the higher-energy irradiations errors from this cause should not exceed 10 per cent, while for the lower-energy irradiations the error should be less.

PHANTOM CONSTRUCTION

A phantom of the thoracic and upper abdominal section of the body was constructed of mix D wax[20] with lateral and sagittal dimensions 30 and 23.5 cm, respectively. For simplicity of construction, the cross-section was made elliptical. All bones lying between T1 and L2 were inserted. The bones were filled with mix D wax in a vacuum oven. Lung tissue was simulated by a cork and wax mixture of density 0.3 g/ml. Fig. 3 shows the phantom with an irradiation cell being inserted.

EXPOSURE CONDITIONS

Dosimeter cells were inserted into close-fitting

cavities in (a) T7 and T8, the interfaces of which were ground slightly to obtain a flat fit, and (b) the sternum. The phantom was irradiated in the fields listed below, rotating at 3 rev/min.

(a) 50 kVcp, unfiltered; 25 mA for 375 min; HVL 0.1 mm Al; target to center of phantom 1.5 m; total air dose 20,000 r.
(b) 60 kVcp, 0.5 mm Al filtration; 25 mA for 360 min; HVL 0.65 mm Al; target to center of phantom 1.5 m; total air dose 7200 r.
(c) 80 kVcp, 1 mm Al filtration; 20 mA for 120 min; HVL 1.5 mm Al; target to center of phantom 1.5 m; total air dose 1790 r.
(d) 100 kVcp, 1.5 mm Al filtration; 20 mA for 60 min; HVL 2.1 mm Al; target to center of phantom 1.5 m; total air dose 748 r.
(e) 150 kVcp, 0.5 mm Cu filtration; 15 mA for 45 min; HVL 0.7 mm Cu; target to center of phantom 1.5 m; total air dose 720 r.
(f) 250 kVcp, 1 mm Cu + 0.5 mm Al filtration; 10 mA for 60 min; HVL 1.9 mm Cu; target to center of phantom 1.3 m; total air dose 367 r.
(g) Co^{60} γ; source to center of phantom 1.5 m; total air dose 107 r.

The 150 and 250 kVp irradiations were carried out using a Mueller M.G. 300 constant-potential unit. For the lower energies a Philips type FA-100 tube with beryllium window was employed.

All exposure doses were measured at the position of the phantom center with the phantom removed, using a Baldwin Farmer dosimeter calibrated at the National Research Council. In the case of the 50 and 60 kVp irradiations, this instrument was calibrated against a free-air chamber constructed for spectra below 100 kVp. This calibration, at energies which could be compared, agreed with the NRC calibration to within 5 per cent. All quoted air doses were measured at the position of the phantom center in the field, with the phantom removed.

RESULTS

The energies absorbed in the tissue as measured by the small-volume dosimeter at the two chosen sites are given in Table 5.

Table 5. Energy absorbed in rads per roentgen exposure (rotational), in sternum and thoracic vertebrae of a human phantom, at various photon energies

Exposure fields (rotational)	Energy absorbed; rads per roentgen exposure at phantom centre	
	T_7–T_8	Sternum
Co^{60}	0.80	0.80
250 kVp	0.60	0.60
150 kVp	0.38	0.49
100 kVp	0.19	0.28
80 kVp	0.11	0.19
60 kVp	0.033	0.089
50 kVp	0.004	0.016

MEAN BONE MARROW DOSE

In order to estimate the mean marrow dose received in an exposure, the distribution of the active marrow in the body and the dose at various marrow sites must be known. A literature survey has not revealed this information directly. However, the total marrow volume and weight were determined by MECHANIK[21] and the distribution of red and fatty marrow was described by PINEY[22]. It has been possible to select the bones which contain only red marrow from the latter reference and obtain the marrow weight in each from MECHANIK. Fortunately, the only bones which contain red and fatty marrow are the humerus and femur. In each the amount is relatively small and the error introduced into the calculation of active bone marrow distribution by neglecting these bones is small. Active bone marrow distribution is presented in Table 6. On the basis of this distribution, the mean bone marrow dose, neglecting enhancement due to wall effect, for the different exposure conditions was estimated (Table 7), assuming that 60 per cent of the active marrow received a dose similar to that measured in the thoracic vertebrae and 40 per cent received a dose similar to that measured in the sternum. This data is presented in graphical form in Fig. 4.

Table 6. Distribution of active bone marrow in the human skeleton

Skeletal part	Active Marrow (%)
Cranium and mandible	13
Ribs	15
Scapulae	5
Clavicles	2
Sternum	3
Sacrum	9
Pelvis	23
Vertebrae	30

EFFECT OF BONE ENHANCEMENT and MARROW CAVITY SIZE

Bone marrow tissue in intimate contact with the bone structure experiences an increase in energy absorbed due to the influence of electrons from the region of high-energy absorption. The increase in mean dose in a small tissue cavity is a function of (a) the size of the marrow cavities and (b) the bombarding photon energy. The work of ENGSTROM *et al.*[23] on spongious bone shows that, for human vertebrae, bone-marrow cavities extend in size from 300 to 900 μ with a mean around 550 μ. For spongious human rib the cavities range from 100 to 700 μ with a 350 μ mean. Using these distributions and the theoretical treatment of SPEERS[24] on the increase in energy deposition in small tissue cavities surrounded by bone, an estimate has been made of the mean enhancement of the tissue dose in vertebrae and ribs at various photon energies (Table 8).

Table 7. Estimation of mean bone marrow dose in rads/roentgen exposure (rotational) at various photon energies

Exposure field (rotational)	Effective energy (keV)	Mean bone marrow dose rads/roentgen
Co^{60}	1250	0.80
250 kVp	105	0.60
150 kVp	70	0.42
100 kVp	29	0.23
80 kVp	25	0.14
60 kVp	19	0.055
50 kVp	10	0.009

The values in Table 8 therefore indicate the approximate factors by which the actual marrow dose will exceed the measured dose because the larger dimensions of the dosimeter result in very nearly zero average enhancement for it. There is an additional error due to the finite volume of bone removed for inserting the chamber but the chambers are small and the glass of the walls has electron density comparable to that of bone. Therefore this error is thought to be small and an estimate of its magnitude has not been attempted.

Table 8. Estimation of dose enhancement to bone marrow due to proximity of bone wall in human vertebrae and ribs at different X-ray energies

Effective energy (keV)	Mean enhancement in vertebrae	ribs
200	1.03	1.05
100	1.07	1.09
50	1.10	1.17
35	1.08	1.12
25	1.04	1.06

DISCUSSION

The measured values of absorbed dose in the bone marrow varies by a factor of almost 100, in relation to unit air dose, in going from Co^{60} to 50 kVp unfiltered X-radiation, but the mean enhancement of the bone marrow dose due to the presence of bone does not change much with photon energy. Assuming that for Co^{60} γ's there is no intensification of dose, then the maximum effect is not likely to be greater than 15 per cent at any energy and is small compared to the drastic reduction in absorbed marrow dose due to attenuation by the bony structure.

There is little experimental knowledge on the LD50 for humans at different photon energies, or at any photon energy for that matter. However, ALPEN *et al.*[4] have measured the comparative response of dogs to 250 and 100 kVp X-rays and ALPEN and JONES[5] have investigated the acute lethality to dogs of concomitant 250 and 50 kVp X-rays. Investigations with dogs at higher energies (Co^{60} γ's and 1000 kVp X-rays) have been carried out by ALPEN[25], MICHAELSON[26] and by SHIVELY *et al.*[27]

Table 9. LD50 Exposure dose, mid-line tissue dose and estimated mean bone marrow dose to dogs irradiated at various photon energies

Group	Photon spectrum	Mid-line air dose (r)	Mid-line tissue dose (rads)	Mean marrow dose (rads)
No. 1 of Ref. 5	250 kVp	275	206	165
No. 2 of Ref. 5	250 kVp * + 50 kVp	226 * + 1000	203	145
No. 3 of Ref. 5	250 kVp * + 50 kVp	205 * + 3000	254	150
No. 1 of Ref. 4	250 kVp	281	228	169
No. 2 of Ref. 4	100 kVp	664	316	153
Ref. 24	1000 kVp (330 eff.)	270	235	195
Ref. 25	1000 kVp (450 eff.)	303	250	222
Ref. 26	Co^{60}	376	335	300

* Concomitant exposures.

If it is assumed that the differences in bilateral and rotational exposure and the effects of physical dimensions are small, then the estimated mean bone marrow dose (using the results summarized in Table 8) to the various groups of dogs irradiated by these investigators is as in Table 9.

The correlation at X-ray energies, from 50 kVp to 250 kVp, between LD50 for dogs and estimated mean bone marrow dose is excellent. However the value of the LD50 for 1000 kVp X-rays and Co^{60} γ-rays are higher. The average value for the lower X-ray energy bone marrow doses could probably be increased by 10 per cent, due to enhancement, to a value of 175 rads (Table 8). Thus if the dose to the blood-forming organs is the governing factor when acute lethality is considered then RBE values are 0.6, 0.8 and 0.9 for Co^{60} γ's, 1000 kVp (450 keV eff.) and 1000 kVp (330 keV eff.) respectively, relative to 250 kVp X-rays. A difference in RBE correlated to the difference in LET between 250 kVp X-rays and Co^{60} γ-rays has been observed previously. STORER *et al.*[28] quote 0.77 and UPTON *et al.*[29] quote 0.71. The biological effect considered is the 30-day LD50 for mice. Further work using this small-volume dosimetric system on large animal phantoms, in conjunction with LD50 studies on the animals, would be of interest.

Finally, it is emphasized that dose intensification in bone marrow near the bone surfaces has been considered small in comparison to attenuation for acute radiation effects subsequent to bone-marrow damage. Chronic biological effects in bone such as leukemia or bone tumour might be more closely related to maximum dose at a particular site rather than to a mean dose to the bone-marrow tissue. In this case appropriate intensification factors obtained from SPIERS' work[24] could be applied to the measured values of aborbed dose in bone sites, reported in this work, to give the maximum tissue dose to bone tissue interfaces, lacunae, canaliculi and Haversian canals.

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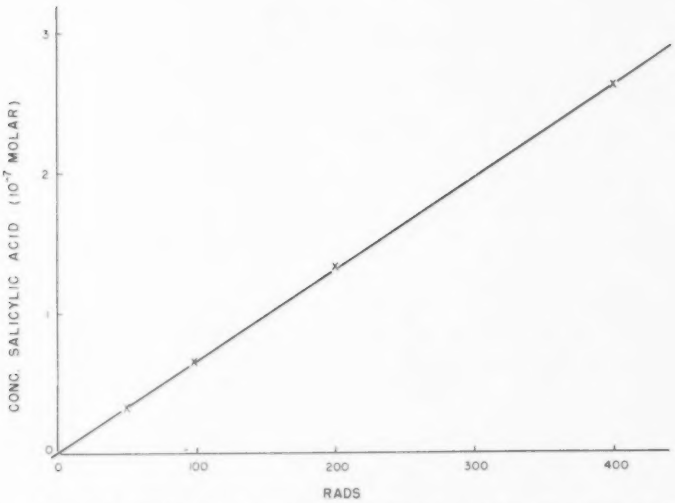

FIG. 1. Linearity of the small volume salicylic acid dosimeter in the 0–500 rad range at 1·25 meV photon energy.

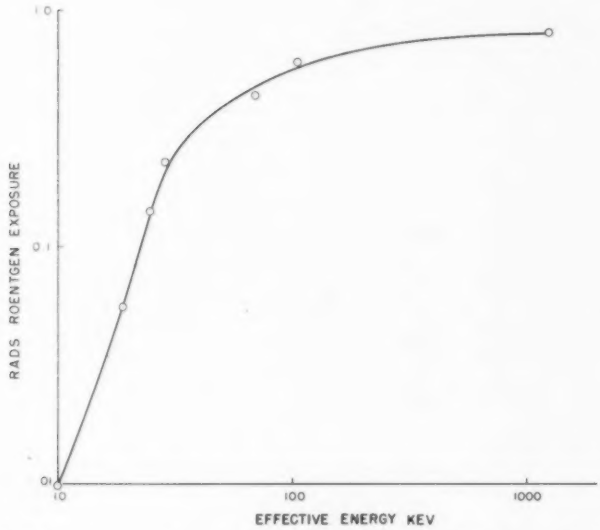

FIG. 4. Mean marrow dose versus photon energy for whole body rotational exposure.

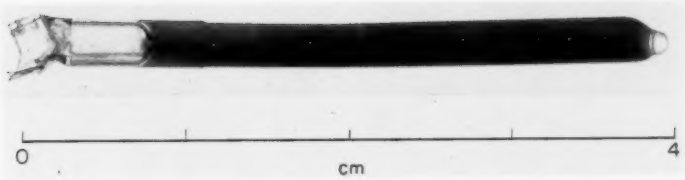

FIG. 2. Irradiation cells.

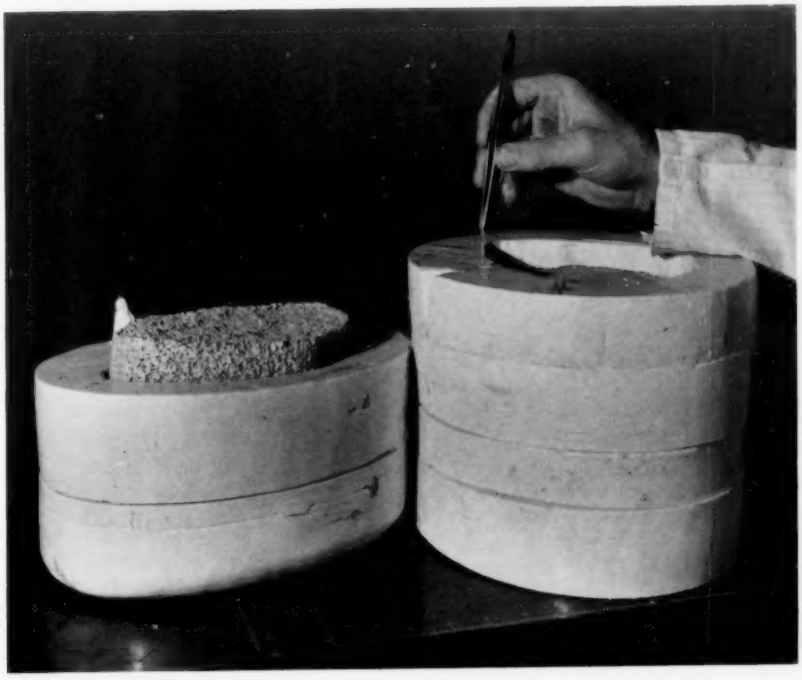

FIG. 3. Irradiation cells being inserted in phantom.

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MIXED RADIATION DOSIMETRY OF A PLUTONIUM–BERYLLIUM NEUTRON SOURCE*

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Abstract—The dose rate delivered to an assumed "standard tissue" by the fast-neutron and γ-radiation from a standard 1 c plutonium–beryllium neutron source was determined by measurements with ethylene-filled polyethylene and CO_2-filled Teflon ionization chambers. The resulting ionization currents as measured with a calibrated vibrating-reed electrometer were used to obtain a measure of both the γ- and fast-neutron components of the emitted radiation. The tissue dose rate due to the fast neutrons extrapolated to 1 cm from the source was found to be 1.74 rads/hr. The γ-component was found to contribute 0.16 rad/hr. From the neutron dose rate and the neutron spectrum for Pu–Be, the emission rate of the source was calculated to be 1.65×10^6 neutrons/sec-c. A discussion of the neutron and γ-spectra of the emitted radiation from this source and some aspects of ionization chamber design are also included in this report.

I. INTRODUCTION

In recent years plutonium–beryllium sources have been coming into much more general use. An advantage over radium–beryllium lies in the fact that Pu–Be has a much smaller γ-component of the emitted radiation. It is also an improvement over polonium–beryllium in that the long half-life of plutonium (24,300 years) effectively removes the necessity of making decay corrections.

When five such sources were obtained at the University of Washington to supply neutrons for a sub-critical reactor, an investigation was initiated to determine the tissue-equivalent dose rate of the emitted radiation. This study was undertaken as an absolute laboratory determination of the dose rate in tissue upon exposure to this type of source and to determine the relative composition of this dose rate resulting from the fast-neutron and γ-components of the absorbed dose.

The biological effect of ionizing radiation is a function of both the energy and type of radiation and that of fast neutrons is about ten times as great as is that of γ-radiation. As there is apparently no neutron source entirely free of a γ-component, a complete analysis of the dosimetry of such a source should, then, properly include not only a measure of the total dose rate, but also an analysis of the relative composition of this dose rate. The Pu–Be sources emit both fast neutrons and γ-radiation (plus α-radiation which is absorbed within the source container); hence this becomes a problem in the field of mixed-radiation dosimetry.

By using the values of the neutron emission rate as determined by the source manufacturer[1] and the neutron spectra for such a source[2] combined with the use of constants to convert to tissue dose rate previously determined by others,[3,4] it is possible to estimate the dose rate and its composition. Because of some discrepancies found in the current literature, it was felt that an independent laboratory determination would be more valid and worthwhile.

II. THEORY

The sources used in this project utilize the α-particles from the plutonium in the ${}_4Be^9$ (α, n) ${}_6C^{12}$ reaction to give a copious supply of fast neutrons. The principal source studied was identified by Mound Laboratories† as M-85 and consisted of 15.87 g of Pu (0.98 c) mixed

* This work is a summary of a M.S. Thesis in Nuclear Engineering at the University of Washington, 1958.[24]

† Mound Laboratory, Miamisburg, Ohio.

with 7.94 g of Be to form a complex intermetallic compound $PuBe_{13}$.[1,2,5] Sources M-83 through M-87 were also studied. The 1-c source, as obtained from Mound Laboratories, is encapsulated in a tantalum stainless-steel container 0.90. in by 0.825 in. i.d. and 1.60 in. by 1.00 in. o.d. The complete assembly is welded shut to prevent possible contamination by the plutonium. This assembly comprises the nominal 1-c source (Fig. 1.). Final ownership and control of these sources rests with the AEC because of the fissionable Pu^{239} content. They emit neutrons with an energy spectrum ranging from thermal up to 12 MeV and monoenergetic γ-radiation of 4.4 MeV.

The emission rate as given by Mound Laboratory was 1.63×10^6 neutrons/sec for source M-85.[2]

When a substance is exposed to fast neutrons, the energy absorbed (and consequently the dose delivered to the material) is strongly dependent on the nuclear composition of the material. In particular, hydrogen nuclei, when bombarded by fast neutrons, give rise to rapidly moving protons whose high specific ionization becomes the primary mechanism of energy absorption in the medium. As a result, the dose delivered to a material from fast neutrons is strongly dependent upon the hydrogen content of the material.

In principle, an accurate measurement of the dose delivered to a tissue can be made by using an ionization chamber composed of tissue-like materials. In order to satisfy the requirements of Bragg–Gray theory, the walls of the chamber must be made of materials with the same atomic composition as the assumed tissue surrounding an ionization cavity which is filled with a gas having the same composition as the wall material. This has been achieved by FAILLA[6] and ROSSI[7] in their "Tissue Equivalent Chambers". In order to accomplish this, one must define precisely what is meant by "tissue", and the assumption of some form of standard tissue is implied in the use of any such chamber. Work is continuing in the development of other techniques for measuring tissue dose rates when the exposure is due to fast neutrons in a mixed radiation field.[8,9]

It has been shown[10,11] that, based upon certain design conditions, the dose rate delivered to the wall material of a homogeneous ionization chamber can be determined by the application of Bragg–Gray theory utilizing the relation:

$$E_w = SWJ \tag{1}$$

where E_w is the energy imparted to the wall material from the ionizing radiation, S is the ratio of the stopping powers of the wall and the chamber (equal to the ratio of the densities of the gas and wall provided their atomic composition is identical), W is the mean energy expended in the production of an ion pair in the gas, and J is the measured electronic current.

If an ionization chamber is fabricated of "tissue" or a tissue-equivalent material and filled with a gas whose composition is chemically identical to the wall material, we can then obtain an absolute measure of the dose delivered to the tissue by measuring the ionization current and utilizing previously determined values for S and W. Good values of W are available[8,12,13] for most gases, including ethylene and CO_2.

It has been shown[8,14] that, for a specified standard tissue and for a complex neutron spectrum, the energy absorbed *per molecule* of polyethylene is within 2 per cent of that for the assumed tissue. This work has assumed a tissue with a chemical composition similar to that of the "standard man" as described by KINSMAN.[15] This assumed tissue is $(CH_7O_3)_n$. By evaluating a weighted average of the energy absorbed per molecule per neutron over the Pu–Be neutron spectrum[2] and correcting for the difference in molecular weights between the assumed tissue $(CH_7O_3)_n$ and polyethylene $(CH_2)_n$, a relation is obtained to convert the measured dose rate in polyethylene to that in the assumed tissue. The weighted results indicate that

$$P_n = 1.36\, D_n \tag{2}$$

where P_n is the fast neutron dose in polyethylene in rads and D_n is the tissue dose in rads due to fast neutrons.

HURST[14] has shown both experimentally and theoretically that equation (2) holds over a range of neutron energies from 0.2 up to 20 MeV and that polyethylene is tissue-like in its response within experimental error over this range. From the practical viewpoint this result is significant, since ethylene can easily be surrounded by

FIG. 1. Photograph of the 1-c Pu–Be neutron source.

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polyethylene to provide a Bragg–Gray chamber.

As a result of both theoretical and previously obtained experimental results, it was felt that, short of an identical tissue-equivalent chamber (the assumption of which is, of course, strictly an approximation) a polyethylene–ethylene chamber would be the material of choice for such measurements. Such a chamber was used to determine the total tissue dose rate due to both the fast-neutron and γ-radiation from the Pu–Be neutron source.

Determining the composition of the radiation

As a possible improvement over previously used techniques for determining relative dose rate composition by the use of balanced ionization chambers, such as the Chang and Eng chamber[16] and CO_2-filled graphite chambers[17], an attempt was made here to utilize a CO_2 filled Teflon* chamber to determine the composition of the radiation. The relatively small neutron response of such a chamber can be calculated, thus allowing a method for obtaining the γ-component of a mixed radiation field. HURST has calculated the fast-neutron response of a CO_2-filled chamber on the assumption that only the recoil atoms of the gas contribute to the ionization current caused by the neutrons. The results of his calculations have been experimentally verified by SAYEG *et al.*[17] By extrapolating the data obtained by SAYEG to the effective energy of Pu–Be neutrons (4.2 MeV), a neutron response of 21.3 per cent is obtained for a CO_2 filled chamber.

A Teflon walled chamber has an advantage over graphite in that it is free of hydrogen contamination in the wall material and, as Teflon is pure polymerized tetrafluorethylene $(CF_2)_n$, it is effectively tissue-equivalent for γ-radiation.[18] Thus, a calibrated Teflon chamber can be exposed to a mixed radiation field and, if the total dose rate is known, then by subtracting 21.3 per cent of the neutron dose rate from the measured total dose rate in Teflon (assumed for 4.2 MeV neutrons), we can obtain a measure of the γ-component of a mixed radiation field.

From a combination of the two chambers one can obtain the information desired. For the Teflon chamber the γ-component of the tissue dose rate in rads D_γ (for the Pu–Be neutron spectrum) is given by the equation

$$D_\gamma = T - 0.213\, D_n$$

where T is the total measured dose rate in rads in the Teflon chamber and D_n is the fast neutron dose rate in tissue.

The fast neutron dose rate can be determined from the polyethylene chamber readings by the relation

$$P = 1.36 D_n + D_\gamma \qquad (4)$$

where P is the total measured dose rate in rads in polyethylene as determined with the polyethylene chamber. Thus, by using the measurements from the two chambers and solving the above simultaneous equations, a measure of both the total dose rate and its composition can be obtained, based on the assumption that the γ-response of both of the chambers is the same as that of tissue.

III. EXPERIMENTAL METHOD

To obtain a measure of the dose rate in tissue, equation (2) was applied and the dose rate measured in a homogeneous polyethylene–ethylene chamber (Fig. 2). The ionization currents were measured with an electrostatically shielded vibrating-reed electrometer. Calibrations were obtained and cross-checked from a number of sources. The electrometer was calibrated with a potentiometer utilizing precision capacitors and resistors which were borrowed from the staff of the University of Washington 60-in. cyclotron. The response of the chamber was checked by exposure to a calibrated Co^{-60} source. The effective volume of the ionization chamber was determined to be 100 ± 1 cm^3 by measurements and by application of a graphical analysis procedure to account for the effect of the guard rings. By calculation utilizing equation (1), good agreement (± 1 per cent) was obtained upon exposure to the Co^{-60} source and comparison with the measured values of the ionization current and the accepted value of W for electrons in ethylene of 26.4 eV/i.p.†[12] Thus

* DuPont trademark for tetrafluorethylene resins.

† i.p. represents an ion pair produced.

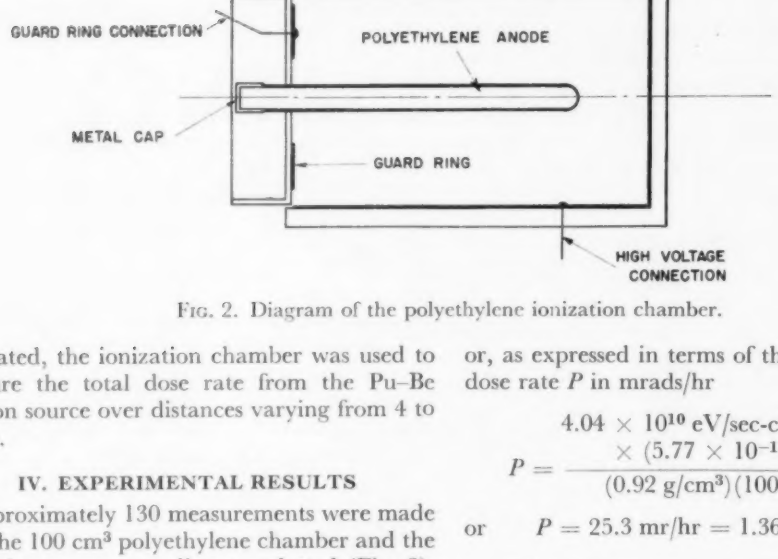

FIG. 2. Diagram of the polyethylene ionization chamber.

calibrated, the ionization chamber was used to measure the total dose rate from the Pu–Be neutron source over distances varying from 4 to 30 cm.

IV. EXPERIMENTAL RESULTS

Approximately 130 measurements were made with the 100 cm^3 polyethylene chamber and the ionization current vs. distance plotted (Fig. 3). From this, an ionization current of $4.6 \pm 0.1 \times 10^{-13}$ coulomb/sec was obtained for an exposure 10 cm from the source. Using this value, the *total* dose rate in polyethylene at 10 cm can be determined by application of Bragg–Gray theory where

$$J = \frac{4.6 \times 10^{-13}\ \text{C/sec-chamber}}{1.592 \times 10^{-19}\ \text{C/i.p.}}$$

$$= 2.89 \times 10^{6}\ \text{i.p./sec-chamber}$$

and S, which is equal to the ratio of the densities of the wall material to the filling gas for a homogeneous chamber, was measured to be 4.98×10^2.

Using the most recently accepted value of W for protons in ethylene of 28.0 eV/i.p.,[12] the energy absorbtion rate in the chamber walls can be obtained from the Bragg–Gray relation [equation (1)]

$$E_w = SWJ$$

or

$$E_w = (498)\ (28.0\ \text{eV/i.p.})\ (2.89 \times 10^{6}\ \text{i.p./sec-chamber})$$

$$= 4.04 \times 10^{10}\ \text{eV/sec-chamber}$$

or, as expressed in terms of the total *polyethylene* dose rate P in mrads/hr

$$P = \frac{4.04 \times 10^{10}\ \text{eV/sec-ch.}) \times (5.77 \times 10^{-11}\ \text{g-rad-sec/hr-eV})}{(0.92\ \text{g/cm}^3)(100\text{cm}^3\ \text{/ch.})}$$

or $$P = 25.3\ \text{mr/hr} = 1.36\, D_n + D_\gamma \qquad (5)$$

FIG. 3. Plot of the ionization current vs. distance.

To separate the γ-component, a CO_2-filled Teflon chamber was calibrated against a Victoreen R-meter and exposed to the neutron source. A total Teflon dose rate of 5.27 mrads/hr at 10 cm was obtained. Using equation (3) we can then write

$$D_\gamma = 5.27 - 0.213\, D_n \qquad (6)$$

By solution of the above two simultaneous equations (5) and (6), the fast neutron (D_n) and γ-components (D_γ) of the *tissue* dose rate from the *0.980*-c Pu–Be source were determined to be

$$\left.\begin{aligned} D_n &= 17.4 \text{ mrads/hr} \\ D &= 1.6 \text{ mrads/hr} \end{aligned}\right\} \text{ at 10 cm}$$

which gives a relative composition of the absorbed dose rate of approximately 92 per cent fast neutron and 8 per cent γ. A reasonable error would be ± 5 per cent for the neutron dose rate and ± 15 per cent for the γ-component as determined from the reproducibility of the measurements and the absolute calibration of the system.

To convert the results into the more useable form, the *tissue* dose rate from a 1-c Pu–Be neutron source at 1 cm will be stated as

$$1.9 \text{ rads/hr} \pm 5 \text{ per cent at 1 cm}$$

with a composition 92 per cent fast neutron and 8 per cent γ.

V. DISCUSSION

Determination of other constants

From the neutron dose rate, the neutron emission rate of a 1-c source can be calculated. Utilizing a recent evaluation of the factor to convert neutron flux to tissue dose rate integrated over the Pu–Be neutron spectrum, which has been evaluated by DEPANGER[4] at Hanford to be 0.0144 mrad/hr-n per cm^2-sec, and making corrections for the thermal neutrons emitted of 5 per cent[2] and fast neutrons removed by absorption in passing through the wall of the chamber by application of fast removal theory[3], the emission rate of a 1-c source was calculated to be

$$1.65 \times 10^6 \text{ neutrons/sec-c}$$

This agrees favorably with the value of 1.60×10^6 obtained by STEWART[2] and the value of 1.66×10^6 calculated from the data provided by the source manufacturer.[1] From this, the "efficiency" of the α-neutron reaction in Be for $PuBe_{13}$ was calculated to be 45×10^{-6} neutrons/α. This agrees with work done by DAINTY[8] in which he reported a value of 45×10^{-6} for Po–Be and with a determination made by HALPERN at Los Alamos in 1949 in which he obtained a final average value of 50×10^{-6}.[19]

Consideration of the γ- and neutron spectra

No presentation on the mixed radiation dosimetry of such a source would be complete without an evaluation of the energy spectrum of the emitted radiation. As for the γ-spectrum, the available information[15,20,21] indicates that there is only one energy of any consequence. This is the 4.4 MeV γ resulting from carbon returning to the ground state following the $_4Be^9$ (α, n) $_6C^{12}$ reaction. This was checked for the Pu–Be source (most available literature on the γ-spectrum is on Po–Be sources) by measuring the γ-spectrum on a scintillation spectrometer using a $3 \times 3''$ NaI (Tl) crystal with a 256-channel analyzer. Only a single γ-energy of 4.44 MeV was observed.

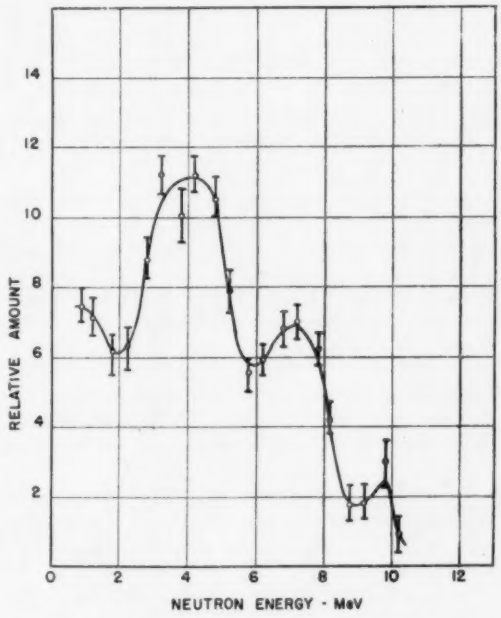

FIG. 4. Pu–Be neutron spectrum.[2]

The determination of the neutron spectrum, however, is a little more complex and enlightening than that of the monoenergetic γ-spectrum. For energies less than about 500 keV, the data are not available (the total amount below this is probably less than 5 per cent). The spectrum of energy distribution above this value has been evaluated by STEWART[2] by the use of nuclear emulsion techniques. Her results, as determined from 2057 observations of nuclear tracks, are shown in Fig. 4. From this spectrum STEWART has evaluated an effective neutron energy of 4.4 MeV and an average energy of 4.2 MeV. The spectrum obtained by her agrees with those obtained for Po–Be[22] and Ac–Be[23]. Thus, as far as the neutron spectra are concerned, a comparison between Po–Be and Pu–Be sources is valid within the currently available data.

VI. SUMMARY

The dose rate resulting from exposure to a 1-c Pu–Be neutron source in an assumed "tissue" (CH_7O_3) and its relative fast-neutron and γ-composition has been experimentally determined. This was accomplished by utilizing a polyethylene–ethylene ionization chamber and a CO_2-filled Teflon chamber to determine the values of the two unknowns in question, i.e. the total dose rate and its composition. The results of this work[24] plus a compilation of other important data on Pu–Be neutron sources are summarized in Table 1.

Table 1. *Data sheet on plutonium–beryllium neutron sources*

Source manufacturer—Mound Laboratories, Miamisburg, Ohio
Source material—2.43×10^4 year $PuBe_{13}$ (intermetallic compound) in a welded tantalum stainless-steel container
Tissue dose rate at 1 cm—1.9 rads/hr-c
Dose-rate composition—92% fast neutron, 8% γ
Neutron emission rate—1.65×10^6 neutrons/sec-c
Neutron energy—from thermal up to 12 MeV (average = 4.2 MeV)[2]
Calculated γ-emission rate—4.2×10^5 photons/sec-c
γ-Energy—monoenergetic at 4.44 MeV (exclusive of X-rays of Pu^{239} decay which are effectively absorbed within the capsule)
Neutron "efficiency"—45×10^{-6} neutrons/α

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IMPROVED MEMBRANE-FILTER AIR-SAMPLING METHODS FOR ENVIRONMENTAL CONTROL

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Abstract—Air sampling is a useful tool for controlling, as well as for analyzing, the environment. The equipment and techniques described were designed to facilitate the collection of localized samples directly at the source of potential contamination. Routine use of membrane filters was simplified by the design of a small, inexpensive filter holder, providing advantages over commercially available units, e.g. reduced size and weight, tight sealing against any filter material without the twist-lock action which often damages filter membranes, and parts exposed to contamination are expendable. Techniques for determining sample collection rates directly from vacuum-gauge readings are described. A combination α–γ scintillation detector assembly, designed specifically for counting membrane-filter samples, was constructed. Sample preparation techniques for use with internal proportional counters are also mentioned.

I. SAMPLE COLLECTION

SAMPLING is an art as well as a science. It requires perception as well as precision; selectivity in addition to sensitivity. With regard to environmental contamination, we should not only detect but also protect; we should regulate as well as evaluate. Air sampling can be an effective tool for environmental control, especially when applied directly at the potential source of contamination.

Fig. 1 shows a typical air-sampling operation, using a Filter Queen sampler† and HV–70 type filter paper.‡ Large-volume samplers of this type are excellent for detecting and measuring contamination in the general environment, but they are awkward to adapt for sampling from a specific point. Also, while HV–70 paper permits good quantitative counts of radioactive contaminants, some qualitative analyses are difficult, if not impossible, to perform, owing to the composition of the paper. Chemical filter papers are more suitable when chemical analysis may be involved, but are dependent to too great an extent on flow rates to achieve high efficiency.[1–3] Impactors, thermal precipitators, scrubbers, etc., are limited by the physical and chemical properties of the contaminants, and by their low collection rates.

The problem which confronted us was to adopt a sampling system which could be used discriminately to sample sources of potential contamination, and which could be implemented on a rather wide-spread scale in a short time. We wanted a system which would provide versatility both in sampling and in analyzing; one which would be simple to calibrate and operate; and one that would be inexpensive to acquire and maintain.

Membrane filters were chosen as the most suitable medium for the types of sampling anticipated, because of (1) their high efficiency for a wide range of particle sizes, (2) their negligible ash content, (3) the option of microscopic examination of the sample, (4) the low absorption of α-activity and (5) the relatively small effect of dust loading on flow resistance.

Fig. 2 illustrates the same hood air-sampling operation, but utilizing the sampling method which was finally selected. The principle of

* Operated by the University of Chicago for the United States Atomic Energy Commission.

† A Filter Queen vacuum cleaner modified to draw air through a filter paper at a flow rate of approximately 8 ft³/min.

‡ Manufactured by Hollingsworth and Vose Company, West Groton, Massachusetts.

localized sampling is demonstrated by the location of the two small filter holders, called "Mini-Heads". The one at the face of the hood samples the breathing zone of the worker. The second Mini-Head, inside the hood, is used to assess the contamination potential of the experimental process. On the left is the vacuum pump which draws air through both Mini-Heads. The method and equipment were chosen to satisfy two specific requirements: (1) the need to collect air from specific points of interest and (2) the need to collect several simultaneous samples in the vicinity of a given experiment.

The following criteria for development and/or procurement of the needed equipment were determined from the anticipated usage. The filter holder must be (1) leak-proof without damaging the membrane filter, (2) physically small and lightweight, (3) adaptable for in-line sampling and (4) inexpensive (both initial and upkeep). The vacuum pump must be (1) portable, (2) capable of collecting up to three samples simultaneously at nominal flow rates of 1 ft^3/min each and (3) inexpensive (both initial price and maintenance).

A search of the available commercial filter holders failed to produce any which met all of the requirements, so the "Mini-Head" was devised. To meet the requirements of size, weight and cost, the unit was injection molded of polystyrene, and both halves are identical so that only one mold is required. The unit was molded without air-inlet holes primarily as an economic measure, but the necessity of drilling holes provided an opportunity for experimenting with various hole sizes and the possibility of incorporating a flow-controlling orifice as an integral part of the holder. Two assembled Mini-Heads are shown in Fig. 3.

Assembly of the Mini-Head is shown in Fig. 4. To eliminate O-ring seals and the twist-lock action which often ruptures the filter membranes, a flat gasket seal is used. This is found to seal very effectively against most filter materials if they are cut to the proper diameter. The determination of filter diameter to use was largely determined by the readily available sizes of membrane filters, and by the anticipated use of internal proportional counters for analysis. The unit was therefore designed to take 47 mm membrane filters as the optimum size. Smaller filters would reduce the nominal flow rate below 1 ft^3/min, while larger filters would not fit the standard 2-in. planchets to be used in the proportional counter chamber. Chemical filters and HV–70 may be punched out to about 47 mm diameter for use in the holder.

The filter membrane is supported by a porous, stainless-steel disk, $\frac{1}{16}$ in. thick. Use of this material instead of wire mesh allows accurate machining to specified dimensions and eliminates the flexing which occasionally strains the membrane to the point of rupturing at the edge of the gasket seal. Although many suitable porous materials are available, stainless-steel facilitates decontamination if necessary. However, the support disk is protected from contamination by the filter itself, and is normally salvaged whenever a Mini-Head is discarded.

The entire filter holder is easily washed in water or alcohol for decontamination. However, discarding the unit would be less costly than the time involved in decontaminating an ordinary holder. This fact makes the use of the Mini-Head desirable whenever the probability of contamination is serious, such as inside a high-level cave, glove box, etc.

Ordinary rubber bands are used for clamping together the two halves of the Mini-Head. With the flat gaskets, very little pressure is required to produce a tight seal. Using two or three bands insures against breakage.

Prices of the Mini-Head components (as of May 1961) were as follows:

One porous stainless-steel disk—$4.48 (each in lots of 25)

Two gaskets—$0.14 ($0.07 each in lots of 250)

Two plastic pieces—$0.40 ($0.20 each in lots of 250)

Mold—$1.00 ($500 amortized over 1000 pieces)

Detailed specifications for the Mini-Head are available on request.[4]

2. FLOW-RATE CALIBRATION

Fig. 5 illustrates some of the results of tests with various sizes of air-inlet holes and orifices. We found that the flow rate would have to be limited to about 0.6 ft^3/min to maintain a constant flow when operating two or three samplers from one of the small vacuum pumps

FIG. 1. Air sampling with a Filter Queen.

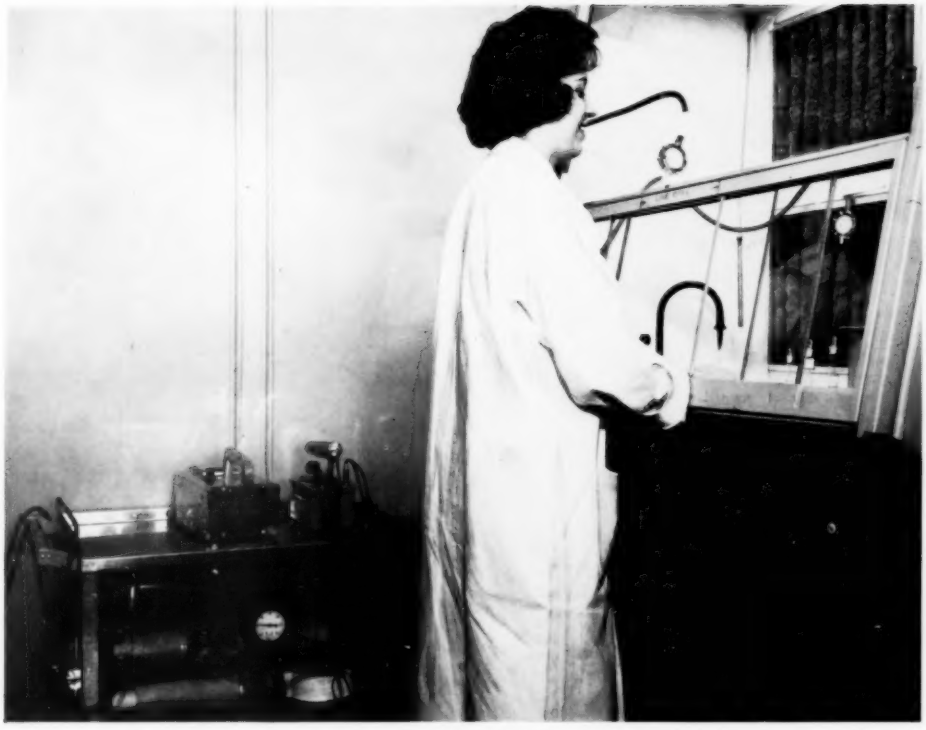

FIG. 2. Air sampling with "Mini-Heads".

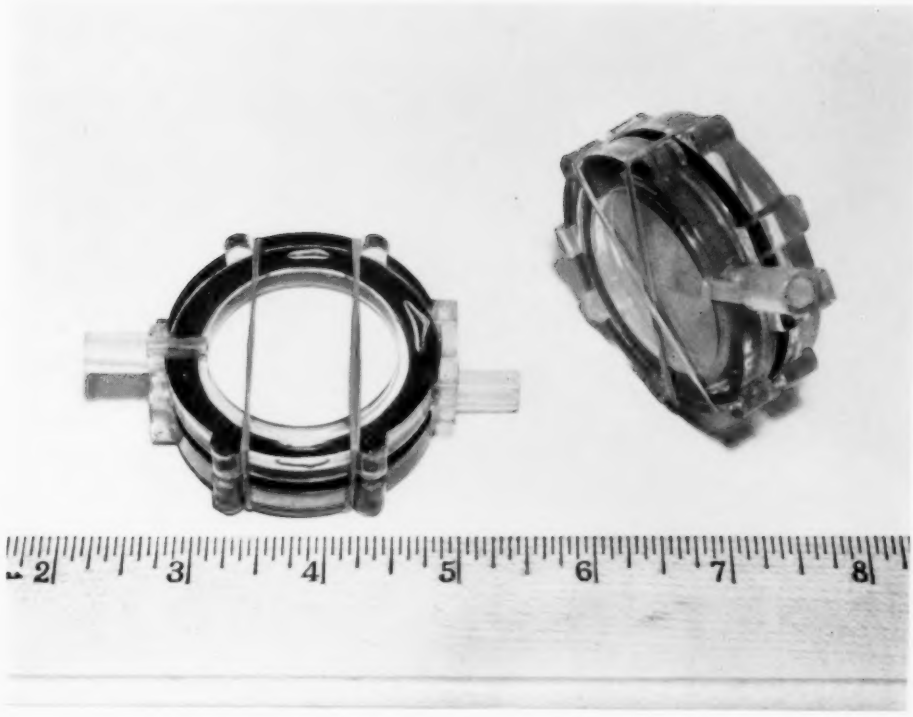

FIG. 3. "Mini-Head" membrane filter holders.

FIG. 4. "Mini-Head" filter holder—exploded view.

FIG. 7. α–γ scintillation counter—disassembled.

FIG. 8. α–γ scintillation counter—assembled.

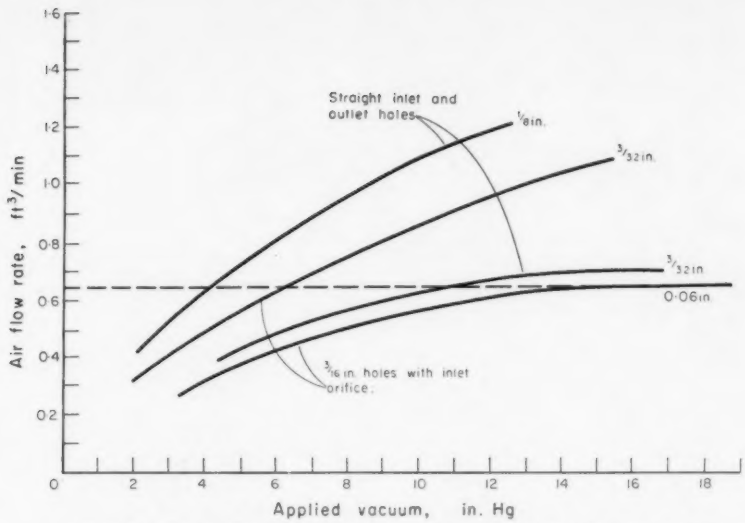

FIG. 5. Effect of hole sizes and orifices on "Mini-Head" air-flow rates—air flow as vacuum.

which we planned to use. We preferred not to restrict the sampling rate so severely, although the use of larger-capacity pumps might make this method of flow control more attractive. Vacuum pumps currently used with the Mini-Heads have free flow ratings of from 3.2 to 6.5 ft³/min, and about 28 in. Hg as the maximum vacuum.

Aerosol distribution on the filter was also investigated for various inlet hole sizes. A $\frac{1}{16}$-in. inlet hole permitted a flow rate of only 0.7 ft³/min, but created a very uneven distribution of dust on the filter. However, a $\frac{3}{16}$-in. inlet hole accommodated three times that flow rate at about one-third the velocity, and non-uniformity of deposition was negligible. As a result, the largest practical hole size ($\frac{3}{16}$ in.) was adopted to achieve maximum flow and uniformity of deposition. Also, since orifice flow control was not practical for our use, we decided to calculate flow rate directly from a vacuum-gauge reading.

The effects of vacuum manifold arrangements and long lengths of rubber tubing on flow rates were also checked. To eliminate discrepancies in flow rates due to manifold design, it was determined experimentally that the manifold should be symmetrical. With equal inlet tube lengths, variations of as much as 15 per cent were found in flow rates in asymmetrical manifolds, the magnitude of the discrepancy depending on the size of the manifold tubing and the separation of the inlet ports.

A very simple symmetrical manifold for connecting up to three filter holders is easily constructed from a $\frac{1}{4}$-in. iron pipe cross. A hole is drilled and tapped ($\frac{1}{4}$-in. I.P.T.) in one side, perpendicular to the original cross inlets. The cross is coupled from this hole to the vacuum-pump inlet by a short $\frac{1}{4}$-in. iron pipe nipple. A vacuum gauge (0–30 in. Hg) is attached in a vertical position to one inlet of the cross, and hose connector nipples for $\frac{1}{4}$-in. i.d. tubing at the other three cross inlets provide the connections to the filter holders. Flow restriction, as well as cost, is reduced by eliminating the use of petcocks at the manifold. Inlets not in use are merely closed by a short length of tubing and a tube clamp.

Pressure drop in the tubing is a function of several variables, e.g. viscosity and density of the air, diameter and length of the tubing, and velocity of the air in the tubing raised to the nth power, where n is also a function of the velocity.[5,6] Although resistance may be computed if these parameters are known, a simpler

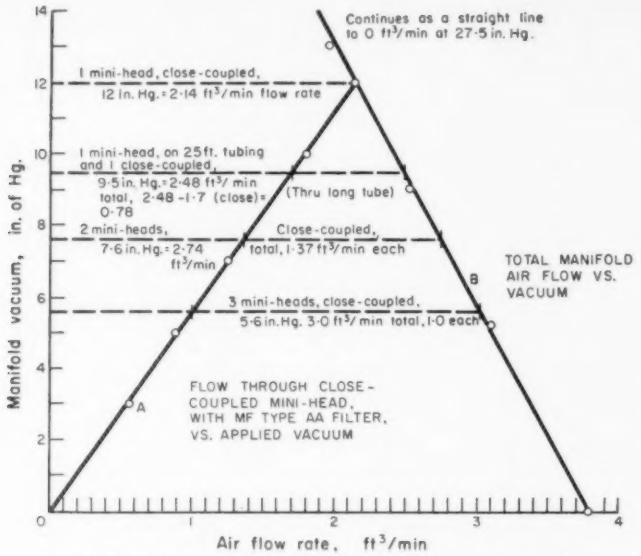
Continues as a straight line to 0 ft³/min at 27·5 in. Hg.
1 mini-head, close-coupled, 12 in. Hg. = 2·14 ft³/min flow rate
1 mini-head, on 25 ft. tubing and 1 close-coupled, 9·5 in. Hg. = 2·48 ft³/min total, 2·48 - 1·7 (close) = 0·78
(Thru long tube)
2 mini-heads, 7·6 in. Hg. = 2·74 ft³/min
Close-coupled, total, 1·37 ft³/min each
TOTAL MANIFOLD AIR FLOW VS. VACUUM
3 mini-heads, close-coupled, 5·6 in. Hg. 3·0 ft³/min total, 1·0 each
FLOW THROUGH CLOSE-COUPLED MINI-HEAD, WITH MF TYPE AA FILTER, VS. APPLIED VACUUM
A
B
Manifold vacuum, in. of Hg.
Air flow rate, ft³/min
0
2
4
6
8
10
12
14
1
2
3
4

FIG. 6. Typical "Mini-Head" flow rate calibration.

method was desired for routine calculations of flow rate when collecting air samples. Calculation of flow rate directly from the vacuum manifold gauge reading is easily accomplished using a graph such as Fig. 6.

Flow rate through a single Mini-Head as a function of the applied vacuum is represented by line *A*. With no connecting tubing, this plot is a straight line, as shown, indicating that the resistance due to the filter itself is directly proportional to flow rate. In actual use, connections made with less than 5 ft of tubing are considered "close-coupled", as the resulting error is negligible. Line *B* represents the maximum capability of the pump for which this graph was made. The intersection of *A* and *B* gives the maximum flow rate possible through a single Mini-Head using MF Type AA membrane filters*, or 2.14 ft³/min in this case. Results typical of the use of multiple combinations of Mini-Heads are indicated on the graph by dotted lines. The only restriction on this method of flow measurement is that any long lengths of connecting tubing must be of equal length. Flow rates will then be equal through all of the filters connected by long tubes, regardless of the location of the Mini-Head in the line. Working within this one limitation is considered preferable to using flowmeters, for the majority of situations encountered.

3. SAMPLE ANALYSIS

As previously mentioned, membrane filters are suitable for many kinds of sample analysis. Ideally, the method of analysis should: (1) require no sample preparation, (2) give both quantitative and qualitative results, (3) be consistent within the statistical fluctuations of radioactive decay, (4) preserve the sample for subsequent analysis by other methods if desired, (5) provide results immediately after sample collections, (6) eliminate all background counts, (7) provide an efficiency of 100 per cent and (8) utilize readily available, inexpensive equipment.

Since all of these criteria have not as yet been met by a single analyzer, a compromise must be reached to best meet the requirements of the specific application at hand. When sampling for purposes of environmental control, time is a very important factor, to facilitate making changes in the operation being monitored. Immediate detection and identification of

* Millipore Filter Corporation, Bedford, Mass.

airborne contamination is, therefore, of the utmost importance. Other considerations are secondary.

For purposes of immediate identification, pulse-height analyzers are desirable, and scintillation detectors are highly suited to such analysis. Efficiencies of 50 per cent or more can be attained quite easily with liquid scintillators, but the sample cannot be easily analyzed by other methods later. Solid crystal scintillators were chosen as the best detectors for routine monitoring use, and have been used initially with standard scalers until pulse-height analyzers could be procured.

To obtain the quickest analysis of the sample, as well as to eliminate unnecessary handling of "hot" samples, we planned to count for α- and γ-activity simultaneously. We preferred to separate the α- and γ-pulses physically by using separate detectors, rather than electronically in the counting equipment. The main reason for this was to keep the equipment simple and allow complete interchangeability of scalers or other counting equipment.

The detector and shield assembly, as finally constructed, is shown in Figs. 7 and 8. The detectors are suspended in the central cavity, the α-crystal facing down from the top and the γ-crystal facing upward directly under the filter. The filter is supported by a screen recessed into the slide bar. The top plates, slide and slide spacers are machined from $\frac{3}{8}$-in. steel plate. A minimum of machine work is required since there are no elaborate off-sets. The slide is provided with two filter supports to give a single motion loading system, allowing change of one sample while another is being counted.

The shield consists of concentric cylinders with base plates welded in, and open tops. The bottom plate of the lower section is raised from the floor, and a cable-access hole provided through the outer wall. The anular shield cavity may be filled with any suitable shield material. The sections of shielding shown in Fig. 7 are lead cylinders made from discarded horizontal Geiger–Mueller tube counting shields. The remainder of the space is filled with lead shot. The upper section does not need to be completely filled to provide adequate shielding for the γ-crystal. Detailed specifications for the shield assembly are available on request.[7]

At present, a $1\frac{1}{2}$-in. diameter by 1-in. thick sodium iodide crystal is being used as the γ-detector. Zinc sulfide in the form of a decal* serves as the α-crystal. Both are mounted so that the crystal just clears the slide. Currently, with the shielding provided, the γ-background is about 150 counts/min with a yield† of 8 per cent for γ's of 1 MeV. The α-background is 0.3 counts/min, with a yield of 18 per cent for normal uranium and 31 per cent for Pu^{239}.

Because of their availability, internal proportional counters have been extensively used for counting membrane filter samples. For this, a quick and convenient method of sample preparation was needed to eliminate the effect of space charges during counting. There are several methods of doing this, but our best results were obtained using a suspension of aquadag (colloidal graphite) in distilled water, with a very small amount of liquid detergent added as a wetting agent. Preparation is accomplished by placing the filter in a cupped planchet containing the aquadag solution. A dropper bottle was found to provide the simplest method of applying the liquid, the quantity of solution used apparently being more critical than the composition of the solution.

4. SUMMARY

Mini-Heads are now used routinely for sampling from hot lab. cells, glove boxes, and other enclosures. Although such sampling may appear unrelated to health protection, valuable information can be obtained regarding process controls, particle-size distributions, filter efficiencies, etc. The small size of the Mini-Head and connecting tubing and the limited sample volume are distinct advantages when sampling such highly contaminated atmospheres. Access to the interior of an enclosure is more easily provided for small tubing than for large sizes; the volume of contaminated materials requiring disposal is kept to a minimum; and the activity levels on the samples themselves are more easily controlled.

* Har-D-Cal Company, Williston, South Carolina.

† "Yield" here denotes the product of detector efficiency and geometry factor.

Sensitivity of any air-sampling system does not depend solely upon the volume collected, but on several factors, e.g. the number and location of samples collected, and the versatility and efficiency of the analysis. In this respect, collection of several membrane filter samples, counted with internal proportional counters or scintillation spectrometers, provides sensitivity superior to high-volume samplers for many applications.

The cost of any sampling system is an important consideration, although secondary to that of providing sufficient, accurate information for evaluation. Current costs of the Mini-Head components have already been mentioned, and may be compared to advantage with other available types. Vacuum pumps with free flow capacities of 3–6 ft^3/min are priced in the range of \$85–130, which is about equal to some, and much less than most, high-volume samplers. The filter papers themselves are a continuing expense in an air-sampling program. The 47-mm dia. membrane filters can be purchased for \$14 per 100, compared with prices of \$4 per 100 and up for various high-volume filter papers. For a program involving, for example, 2000 samples per year, the increased cost of the filters would be \$200 annually.

It is anticipated that the number of small, localized air samples, collected on membrane filters, will continue to increase. As a result, more detailed and accurate information about specific breathing zones and the contamination potential of various processes will be made available. More accurate information will make the Health Physicist's job easier, by inspiring greater confidence in his hazard evaluations and recommendations for environmental control.

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RADIOACTIVITY DISTRIBUTIONS IN THE METALLURGICAL PROCESSING OF THORIUM

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Abstract—The radioactivity distribution patterns produced in the metallurgical processing (particularly casting by consumable arc melting) of thorium metal have been studied. Samples from various locations in the metal, from material deposited on process equipment, and from process area environmental sampling were subjected to gross counting, α- and γ-spectrometry, and growth-decay studies. Separation of thorium and radium and the release of thoron gas are the two important effects observed. A simple γ-spectrometry photopeak ratio method for determining the enrichment or depletion of radium in thorium samples, along with its calibration, is described. The data indicate extensive removal of the radium daughters from thorium in arc-melting, two successive arc-meltings removing essentially all of the Ra^{228}. The radiological hazard and analysis implications of these radioactivity distribution patterns are discussed.

INTRODUCTION

An opportunity to study the radioactivity distribution occurring in the metallurgical processing of thorium was provided in an experimental program involving the casting by consumable arc melting of thorium metal. Variable distributions of radioactivity were expected from thorium processing because of the several chemical entities involved and because of the half-lives of the members of the Th^{232} decay series. The primary interest in these distribution patterns was generated by concern over the radiological hazards involved in processing thorium and any increase in those hazards because of process-induced changes in the radioactivity distribution. The possibility of releasing high specific activity daughter products from the low specific activity thorium system was of particular interest.

Gamma- and α-energy spectrometry were considered ideal tools for studying the radioactivity patterns, with γ-spectrometry being of particular interest for use on as-received samples of various physical configurations.

* Operated by Union Carbide Nuclear Company for the United States Atomic Energy Commission.

RADIOACTIVITY OF THORIUM-232 SERIES

At the initiation of the studies on thorium, a discussion of the radioactive properties of thorium and the implications of variations in the decay chain equilibria was prepared as a guide for technical persons involved in the processing of thorium or in the radiological health monitoring of thorium processing.[1] It is necessary to summarize briefly that discussion in describing the analytical methods applied and the radioactivity patterns observed.

The term "thorium" is generally used to refer to the isotope Th^{232}, which occurs in nature with an isotopic abundance usually described as 100 per cent, and its daughters. Th^{232} is the parent of the thorium or $4n$ series, a natural radioactive decay chain, which is shown in Fig. 1.

In naturally occurring thorium minerals, all of the decay chain shown in Fig. 1 will be present. Unless the natural state has been disturbed, the nuclides will be in secular equilibrium with the activity of each daughter being equal to the activity of the parent Th^{232}. Thus, in Fig. 2, the α-spectrum of a sample of thorium indicates an α-peak for each α-emitting

FIG. 1. Decay scheme diagram for Th^{232} (thorium or $4n$) series.

member of the thorium series. The trace of Th^{230} shown is the daughter of uranium originally present in the mineral. Th^{230} is frequently found with Th^{232} in such samples. The amplitudes of the peaks compared to that for Th^{232} are approximately equal to the α-activity ratios attributed to each member in the decay scheme of Fig. 1.

Important to this study is the fact that the $4n$ series contains two thorium isotopes, Th^{232} and Th^{228}, each the parent of a radioactive sub-series. The Th^{232} and Th^{228} will not be separated by ordinary chemical or physical treatment. The significant decay chain disruption which can be caused is the partial or complete separation of thorium and radium. The daughters of the radium isotopes are so short-lived that whether they remain with the radium or thorium is not important. The Ra^{228} and Ra^{224}, once the chain is disrupted by the removal of the Th^{228} link between the two, may be treated as parents of two separate sub-series. These sub-series behave independently of each other until more Th^{228} can be produced to make the Ra^{224} dependent upon the Ra^{228}.

Since the thorium isotopes would not be separated by normal treatment, the activity ratio of Th^{232}/Th^{228} will be the same immediately after the radium-removing processing and will then gradually increase as the Th^{228} decays. The Ra^{224} sub-series, controlled by the 3.64-day half-life of Ra^{224}, will fairly quickly grow back into equilibrium with the Th^{228} and will then follow the Th^{228} activity. This growth to equilibrium will have reached about 90 per cent of completion after 10 days.

The Th^{228} activity, decreasing due to decay, will also be in a process of growth from the build-up of the Ra^{228} and Ac^{228}. The total Th^{228} activity will decrease for a while before increasing, since the rate of formation (limited

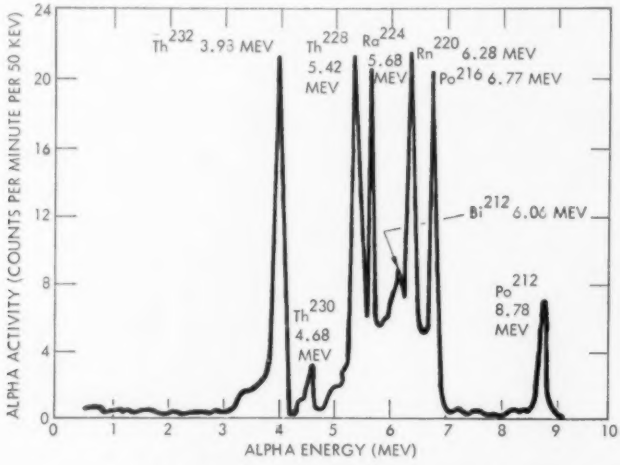

FIG. 2. α-Spectrum of Th^{232} series at secular equilibrium.

by the 6.7-year Ra^{228} half-life) would be lower than the rate of decrease (determined by the 1.9-year half-life of Th^{228}). If the two rates of change and their sums are calculated and graphed for a natural thorium sample after its first purification from radium, the graph of Fig. 3 is obtained. If the radium-removal process is repeated periodically, the Th^{228} activity would be gradually reduced below the indicated minimum.

Fig. 4 is the α-spectrum of a thorium sample which has undergone purification from radium, resulting in Th^{228} and daughter activities which are much lower than the Th^{232} activity.

These changes in Th^{228} content occur gradually, as indicated by the time scale of Fig. 3. In a short period of time, the relative behavior of the Ra^{228} and Ra^{224} indicate radioactivity distribution changes more quickly than the Th^{232}/Th^{228} ratio, because of the short half-life of Ra^{224} and can be indicative even if no thorium remains in a sample.

In a daughter-depleted fraction, such as that described by Fig. 3, the Ra^{224} sub-series will grow back quickly, while the Ra^{228} ($T_{1/2} = 6.7$ years) will not return quickly. As a result, the fraction, about 10 days after depletion, will show a low Ra^{228}/Ra^{224} activity ratio.

In samples containing Th-free daughters or even an enrichment of radium daughters, the Ra^{224} not maintained by Th^{228} will decay through several half-lives before the Ra^{228} decay will be detectable. Thus after about 10 days the Ra^{228}/Ra^{224} ratio will be higher than that at equilibrium.

Another aspect of the thorium series requiring attention is the presence of the inert gas Rn^{220}, classically called "thoron", in the decay chain. It is to be expected from the physical properties of this nuclide that any treatment or handling of thorium may produce some loss of thoron from the material mass. Although thoron itself has a very short half-life, larger than normal amounts of the thoron daughter products, controlled by

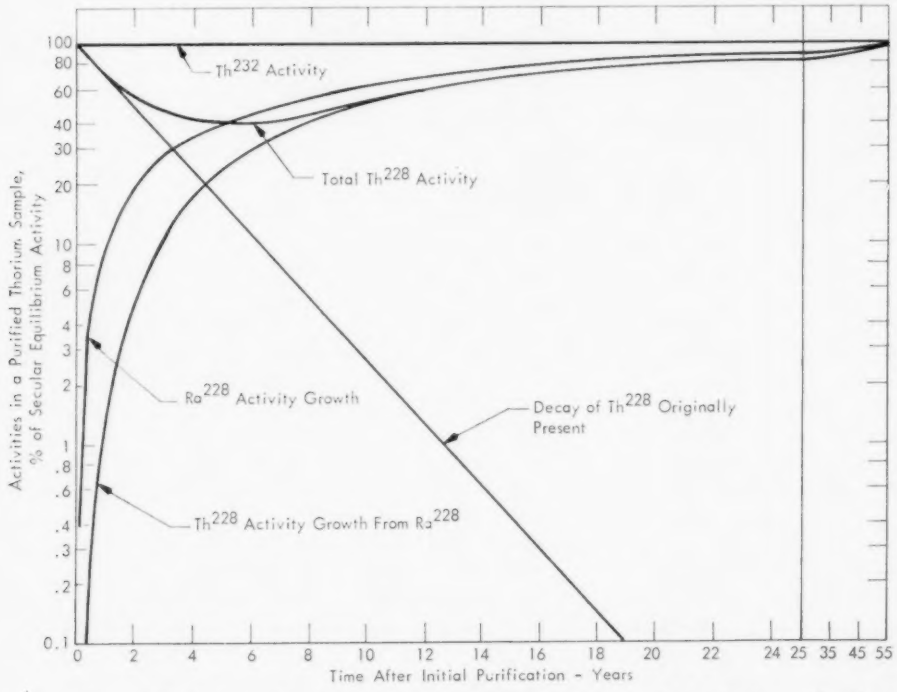

FIG. 3. Variations of activity in a natural thorium sample after a single purification in which all radium is removed.

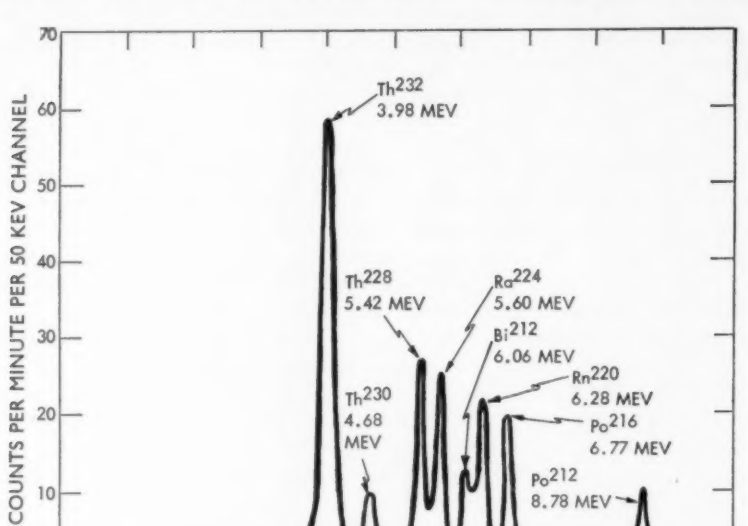

FIG. 4. α-Spectrum of thorium sample after several purifications.

the 10.6-hr half-life of Pb^{212}, can be expected in samples of airborne particulates from atmospheres in which thorium is being processed.

NUCLEAR RADIATION SPECTROMETRY EQUIPMENT

Most of the data were obtained from the energy spectra of the α- and γ-radiation from samples. γ-Spectrometry was carried out with a 1½ in. by 1 in. thick NaI scintillation crystal and 2-in. photomultiplier assembly. α-Spectrometry was performed in a Frisch grid ionization chamber on samples evaporated on stainless-steel planchets.

The voltage pulses from either the γ- or α-detectors are sorted according to pulse height by a 200-channel pulse-height analyzer. This instrument, of the magnetic core memory type, can sort a pulse height or energy distribution into 200 sequential energy increments or channels, storing up to 65,535 events per channel. The energy distribution may be displayed on a cathode-ray tube or a strip-chart recorder or may be digitally recorded on punched paper tape and/or printed tape.

Samples were normally subjected to γ-spectrometry as received, the sample bottles simply being placed on the scintillation detector. α-Spectrometry samples required considerable preparation, the final analysis being on 1¼-in. dia. stainless-steel counting planchets in which solutions of the material to be analyzed were evaporated.

PHYSICAL TREATMENT OF THORIUM

The casting of ingots by consumable electrode arc melting was expected to produce a much greater disruption of the thorium decay chain than the other treatments such as acid "pickling", machining, forging or shaping.

In consumable electrode arc melting, the material to be cast is made one electrode of an electric arc gap and the heat of the arc serves to melt the electrode. The opposite electrode is in a water-cooled crucible in which the molten metal solidifies. The consumable electrode is gradually lowered to maintain the proper arc gap, resulting in the melting of the consumable electrode and the casting of an ingot in the water-cooled crucible.[2,3] Volatile impurities tend to move toward the cold walls of the crucible where they may be removed by light machining of the ingot.

In the studies reported here, the arc melting was carried out under vacuums of about 10-μ pressure, the off-gases of the furnace being filtered to remove radioactive material.

In addition, a stainless-steel furnace liner was used around the consumable electrode and down to the crucible level to collect any splatter or condensing vapors. This liner was used to minimize contamination of the furnace.

The consumable electrodes were made up of 1-in.-dia. thorium rods welded together in a cluster of three or of longitudinal segments of thorium ingots welded into a cluster. Electrodes up to 5 ft in length were used. The arc-melting process was repeated on the "first-melt" ingots so that the metal underwent two arc meltings. The first-melt ingots were 12–18 in. long and $5\frac{3}{4}$ in. in dia., and the "second-melt" ingots 22–23 in. long and $7\frac{3}{8}$ in. dia. The ingot weights were about 60–70 kg for first-melt ingots and about 200–220 kg for second-melt ingots.

The other operations on the thorium included "pickling" in nitric acid before melting, after first melting, and after the machining following second melting; machining of first- and second-melt ingots, and various metal-shaping operations at relatively low temperatures compared to the arc-melting temperature. In addition to samples from all of these operations, "smear" samples of material deposited on the furnace liners and a few air samples were obtained for the radioactivity studies. Care was exercised in taking the machining samples to separate turnings from different areas, and particularly from the first cut and second cut on the outside diameter.

γ-RATIO ANALYSIS FOR Ra^{228}/Ra^{224}

Based on a consideration of the decay effects, observation of the Ra^{228}/Ra^{224} activity ratio no sooner than 10 days after the time of any treatment expected to change the radioactivity distributions appeared to be a suitable indicator of the resultant pattern. Of the methods available, γ-spectrometry was chosen. α-Spectrometry alone would not be indicative of Ra^{228} concentration and therefore would not be applicable for samples containing no thorium. The α/β ratios of samples should be somewhat indicative of radium enrichment or depletion, but were not expected to be very sensitive because of the variety of isotopes contributing to the α- or β-count rates. β-Energy analysis, of course, is a rather tedious and time-consuming technique. Of perhaps more importance than any other factor is the fact that either α- or β-analysis would require rather elaborate preparation of the large number of samples of various types. γ-Spectrometry which could be applied to samples without preparation and without critical concern for sample geometry and still yield the Ra^{228}/Ra^{224} ratio would be acceptable. Determination of actual activity levels in the samples was not required to detect the patterns of distribution.

A γ-spectrum of a sample of the "thorium starting material" or thorium metal rods to be treated metallurgically is shown in Fig. 5 by the solid line. The other spectra are from samples of processed thorium. Various γ-lines are identified from tables of thorium γ-activity.[1]

From this spectrum, the 0.9 MeV γ-line of Ac^{228} was chosen to represent the Ra^{228} activity and the 2.6 MeV line of Tl^{208} was chosen to represent the Ra^{224} activity. These lines were chosen because of their prominence in the spectrum and their relative freedom of Compton scatter effects from other γ-lines. As previously stated, the half-lives of the daughters of radium in each sub-series are sufficiently short that the daughter concentrations will be indicative of radium concentration at the end of the 10 days required for the Ra^{228}/Ra^{224} ratio to be meaningful.

On this basis, the area beneath the photopeaks in the energy range 0.825–1.090 MeV was chosen to represent the 0.9 MeV γ-line and the Ra^{228} activity, and the area under the photopeak in the energy region 2.430–2.655 MeV to represent the 2.6 MeV γ-line and the Ra^{224} activity. The ratio of the 0.9/2.6 MeV was taken for every sample and termed the "gamma ratio". For the starting material whose γ-spectrum is shown in Fig. 5, the γ-ratio is 7.8. Therefore, a γ-ratio higher than 7.8 for a sample would indicate radium enrichment, while a ratio lower than 7.8 would indicate a depletion of the radium daughters, relative to the starting material.

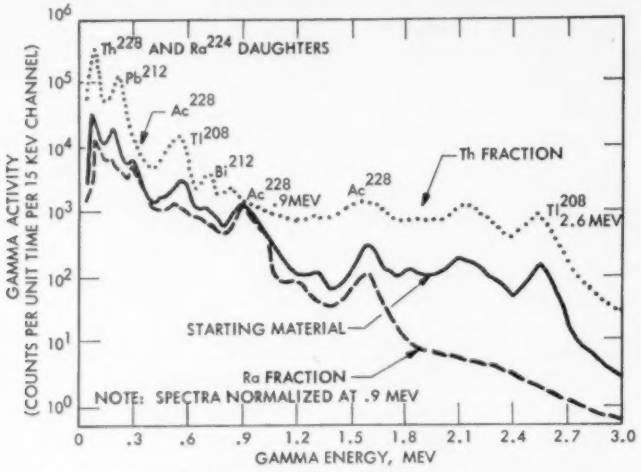

FIG. 5. γ-Spectra from thorium–radium separation experiments.

Although this simple γ-ratio proved capable of indicating the Ra^{228}/Ra^{224} ratio relative to the starting material and of ordering samples according to Ra^{228}/Ra^{224} activity ratio, it was desired to make the analysis as absolute as was feasible and to obtain a calibration of the γ-ratio in terms of actual rather than relative Ra^{228}/Ra^{224}.

To obtain calibration data, a laboratory separation was made to obtain Ra-free thorium and Th-free radium in which the growth patterns could be observed quantitatively. A 5-g sample of the thorium starting material was dissolved in nitric acid containing a trace of hydrofluoric acid, and the thorium was extracted into a solution of tri-n-octylphosphine oxide (TOPO) in cyclohexane.[4]

α-Counting planchets were prepared for each fraction and α- and γ-spectrometry on the thorium and radium fractions were initiated as quickly as possible and repeated at frequent intervals.

The α-spectra of the fractions, taken 4.9 and 2.2 hr after separation, are shown in Fig. 6(a) and (b) for the thorium and radium, respectively. That the separation was essentially 100 per cent is evidenced by the absence of Ra^{224} or other daughters in the thorium spectrum [Fig. 6(a)] and of Th^{232} or Th^{228} in the radium fraction [Fig. 6(b)].

The γ-ratios obtained from γ-spectrometry were tabulated along with the time after separation. In the radium fraction, the γ-ratio increased steadily from the initial value of the starting material and reached a maximum after 24 days. This increase in the first 20–30 days for another sample of this type is shown in Fig. 9. The ratio for the thorium fraction was 1.6 after the same period of time. The γ-spectra of the fractions, from which these ratios were calculated, are the other two spectra of Fig. 5. All of the spectra have been normalized in amplitude at 0.9 MeV for comparison. It may be noted that the 2.6-MeV γ-line of Tl^{208} is absent from the radium fractions, resulting in the high γ-ratio, and that the 0.9 MeV γ-line of Ac^{228} has disappeared from the thorium fraction, causing a low γ-ratio.

Since the variation of the γ-ratio in the radium fraction is due to the decay of the Ra^{224}, the elapsed time since separation for each ratio was converted to the number of half-lives of Ra^{224} ($T_{1/2} = 3.64$ days) and the Ra^{224} activity of the fraction (in per cent of the initial Ra^{224} activity) was calculated. Since the Ra^{224} content is an exponential function of elapsed time, a logarithmic plot yields the straight-line relationship indicated by the solid portion of the curve in Fig. 7. Subsequently, this straight line was extrapolated (dashed portion) to the γ-ratio of the thorium fraction, yielding an effective Ra^{224} content relative to that of the radium fraction.

The Ra^{228} content of the radium fraction, due to the 6.7-year half-life, will have remained

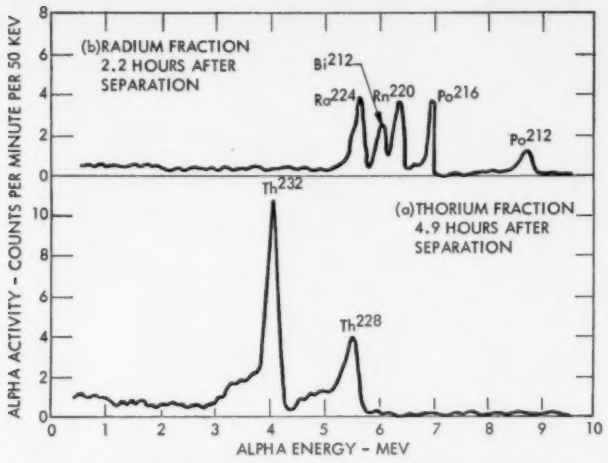

FIG. 6. α-Spectra of Ra-free thorium and Th-free radium fractions.

effectively constant during this 25-day period in which the Ra^{224} activity has varied so widely. If the Ra^{228}/Ra^{224} activity ratio were known for a single point on the graph, the Ra^{224} activity, now expressed as per cent of the initial activity, could be indicated as Ra^{228}/Ra^{224} activity ratio or a series of γ-ratios.

Although the Ra^{228} activity was not known at this time, an assumption was made that the Ra^{228} activity in the starting material, and hence in the radium fraction immediately after separation, was essentially equal to the Ra^{224} activity ($Ra^{228}/Ra^{224} = 1$) in this starting material. If this assumption is valid, a ratio of 1 may be assigned to the point whose abscissa is 100 per cent of initial Ra^{224} activity (radium daughters immediately after separation from thorium). The activity ratios for all other points on the line then become the Ra^{224} activity in per cent divided by 100. This new abscissa has been added to the top of Fig. 7.

The validity of the assumption involved was later proven by a determination of Ra^{228}. It was judged valid at the time of calibration of the γ-ratio method because of the rather high Th^{232}/Th^{228} ratios observed on the starting material and because it was known that the thorium had been in storage for several years. This high ratio (2.23) indicated that the thorium had undergone more separations from radium than the single purification described in Fig. 3. As the relative Th^{228} content becomes lower with repeated purification, it becomes more dependent upon production from new Ra^{228} and less dependent upon its original equilibrium concentration.

Later in the studies, the Th^{228} activity in a sample containing the Ra^{228} which had been separated 263 days previously from 250 μg of thorium starting material (equivalent to 61 dis/min Th^{232}) was determined by α-spectrometry to be 7.7 dis/min. Since consideration of the growth rate of Th^{228} in Ra^{228} indicated that after 263 days the Th^{228} activity would be 21.7

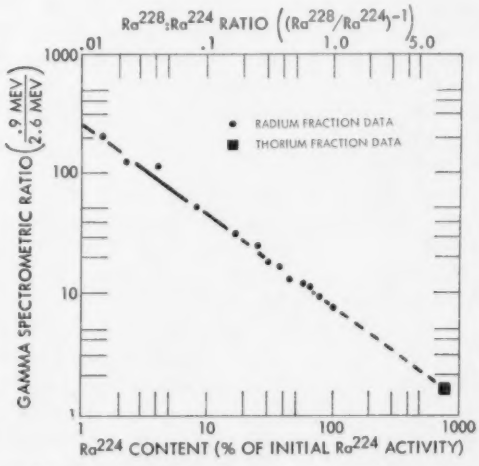
Ra228:Ra224 RATIO ((Ra228/Ra224)-1)
RADIUM FRACTION DATA
THORIUM FRACTION DATA
GAMMA SPECTROMETRIC RATIO (.9 MEV / 2.6 MEV)
Ra224 CONTENT (% OF INITIAL Ra224 ACTIVITY)

FIG. 7. Determination of relationship of γ-ratios to activity by decay studies.

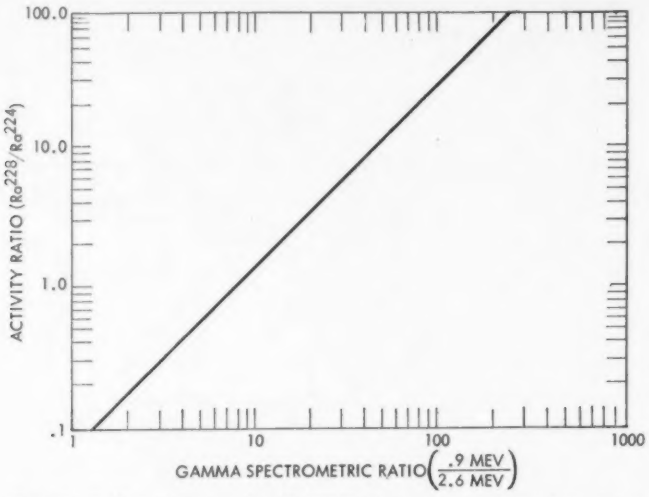

FIG. 8. Calibration graph—Ra^{228}/Ra^{224} ratio as a function of γ-ratio.

per cent of the initial Ra^{228} activity, it was concluded that the initial Ra^{228} activity in the 250 μg of thorium was 26.3 dis/min. Thus the Th^{232}/Ra^{228} activity ratio in the starting material was calculated to be 2.32. This ratio, agreeing quite well with the Th^{232}/Th^{228} ratio of 2.23, means that in the starting material the Ra^{228}, Th^{228} and Ra^{224} all have essentially equal activities.

That the assumption was valid and the starting material Ra^{228}/Ra^{224} ratio is essentially 1 were further evidenced by the observation of the α-spectrum of samples over a period of 300 days. The Th^{228} activity, rather than decreasing showed a very slight increase when compared to the Th^{232} activity. This increase is only possible if the Ra^{228} activity is controlling the Th^{228} activity.

A correlation between γ-ratio and Ra^{228}/Ra^{224} activity having been made in Fig. 7, a more convenient working calibration of Ra^{228}/Ra^{224} activity ratio as a function of γ-ratio was prepared, as shown in Fig. 8.

The γ-ratio method may be used to detect Ra^{228}/Ra^{224} ratio and hence radium enrichment or depletion in thorium samples at least 10 days after the decay chain disruption. It is recognised that, as more time elapses, the slow growth of Ra^{228} will have its effect on the γ-ratio. One sample containing large amounts of apparently Th-free radium was generated in the program and subjected to frequent γ-ratio analysis. The readings on this sample over 320 days are recorded in Fig. 9. It may be noted that after about 35 days, the γ-ratio and hence the Ra^{228}/Ra^{224} ratio begins to drop under the influence of the Ra^{228} decay and production of Th^{228}.

The most attractive feature of the γ-ratio method is that it can be applied to samples essentially as-received.

RADIUM–THORIUM DISTRIBUTIONS

The γ-ratio analysis was performed on all samples. Most of the samples were subjected to two γ-ratio analyses at different times for confirmation and to be certain that the Ra^{228}/Ra^{224} relationship had been established prior to analysis. In total, about 250 samples were received, many of which were of the same operations or circumstances. Thus, data may be shown which are from a single, actual operation and also those which are the summarization of many analyses on an operation.

In Fig. 10 is a sectional sketch of a first-melt ingot, with the actual Ra^{228}/Ra^{224} activity ratios indicated at the location from which samples were taken. It will be noted that trends can be observed, with higher radium enrichments being noticed in the first-cut samples than in the

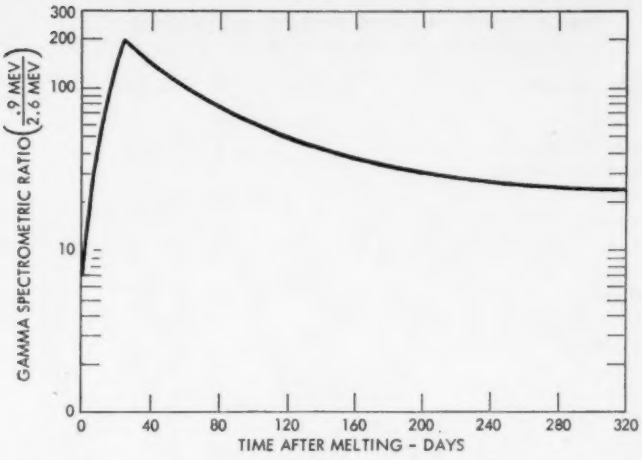

FIG. 9. Long-term behaviour of γ-ratio for a Th-free radium fraction.

second-cut samples. However, there are exceptions to the pattern, such as for sample C-6 in Fig. 10, indicating that the enrichment of daughters in the outer surface was not a completely uniform process.

The distribution patterns are best indicated by Table 1, in which samples from various processes, locations, conditions and ingots are summarized. The number of samples of each condition is indicated, and samples of the same type from various ingots are grouped together to indicate the general uniformity of the distributions observed.

From Table 1, the distribution patterns indicated below are concluded:

(1) The two processes of arc melting remove essentially all of the radium from the interior of thorium ingots. The Ra^{224} and its sub-series, of course, quickly return to equilibrium with the Th^{228}.

(2) In the arc-melting removal of radium, much of the radium in both first and second melting is vaporized, resulting in a deposit of Th-free radium on the furnace liner.

(3) In the ingot itself, on both first- and second-melt ingots, the outermost layers contain the highest radium enrichments, with the second cut layer containing a smaller amount, and the interior the least. In the interior of the ingots, there appears to be a slight gradient, with the top of the ingot being more depleted of radium than the bottom.

(4) The material dissolved from the starting material in "pickling" has a slight radium enrichment, probably because of the last previous casting of the metal. "Pickling" solutions from first-melt ingots before machining contain high radium enrichment. Although second-melt

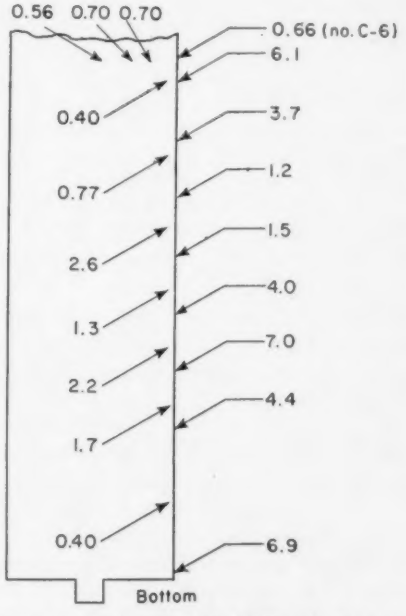

FIG. 10. Location and Ra^{228}/Ra^{224} ratios for samples from a first-melt ingot.

Table 1. *Summary of radium enrichment and depletion data*

Description of sample type	No. of samples	Ra^{228}/Ra^{224} activity ratio
Starting material, rods of thorium	11 analyses	0.98
"Pickle" solution, starting material	1	1.6
Exterior of electrode stub	1	0.125
Interior of electrode stub	1	0.82
"Pickle" solution, 1st melt ingots	1	21.3
1st melt, 1st cut, 1st ingot	9	3.7
1st melt, 1st cut, 3rd ingot	3	2.6
1st melt, 1st cut, 4th ingot	6	3.3
1st melt, 1st cut, 6th ingot	6	7.6
1st melt, 1st cut, 7th ingot	6	6.8
1st melt, 2nd cut, 1st ingot	7	1.3
1st melt, 2nd cut, 3rd ingot	3	3.0
1st melt, 2nd cut, 4th ingot	6	1.15
1st melt, 2nd cut, 6th ingot	6	1.2
1st melt, 2nd cut, 7th ingot	5	0.68
1st melt, interior cut, 1st ingot	3	0.66
1st melt, interior cut, 3rd ingot	1	0.29
1st melt, top interior cut, 4th ingot	3	0.29
1st melt, top interior cut, 6th ingot	3	0.31
1st melt, bottom interior cut, 6th ingot	1	0.62
1st melt, bottom interior cut, 7th ingot	3	1.0
2nd melt, 1st cut, 1st ingot	6	0.74
2nd melt, top interior cut, 1st ingot	3	0.16
2nd melt, center interior cut, 1st ingot	4	0.18
2nd melt, bottom interior cut	3	0.29
"Pickle" solution, machined 2nd melt metal	1	0.17
2nd melt metal, surface after forging	10	0.18
2nd melt metal, final machining	2	0.16
Furnace liner smears, 1st melting, series 1	9	54.0
Furnace liner smears, 1st melting, series 2	9	45.0
Furnace liner smears, 2nd melting	9	43.0
Thorium fraction from laboratory Th–Ra separation	1	0.13
Radium fraction from laboratory Th–Ra separation	1	46.0

ingots were not "pickled" before machining, material dissolved from the second-melt later in the process showed radium depletion, relative to the starting material.

RESULTS OF α- AND β-ANALYSIS

Certain samples of those described above were selected for α- and β-analysis. No more elaborate β-analysis than gross β-counting was attempted, but α-spectrometry as well as gross α-counting was carried out.

In the case of metal turnings, a weighed amount of metal was dissolved in concentrated HNO_3 and a small amount of H_2SiF_6. A 1-ml volume containing 300 μg of thorium was evaporated on stainless-steel counting planchets with a drop of Aerosol-OT wetting-agent solution added to insure a uniform deposition. Smear and filter-paper air samples were charred and ignited at 600°C before being dissolved. Liquid samples were taken directly from the "pickle" solutions for evaporation on planchets.

After the α-spectra had been determined, areas under the α-peaks for various thorium series members were tabulated and compared. Fig. 11 is one of the more interesting spectra,

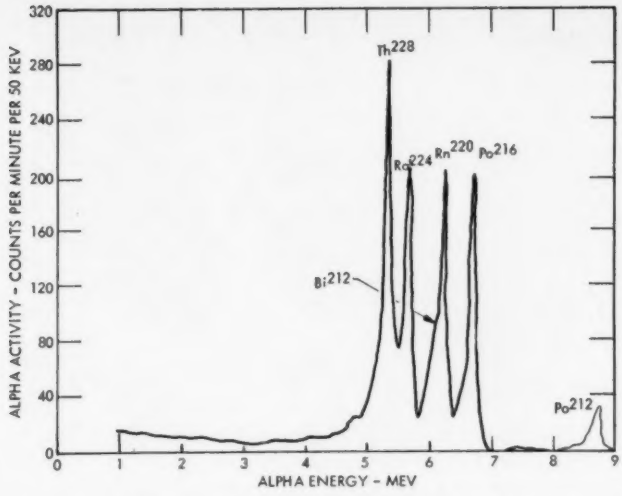

FIG. 11. α-Spectrum of an originally Th-free radium sample from a furnace liner, showing growth of Th^{228}.

showing the Th^{228} sub-series which had grown into the Ra^{228} present in a smear sample after 300 days.

From the gross α- and β-counting in a windowless proportional counter, the α/β count rate ratio was determined.

Table 2 is a summary of the data on the samples selected for this special treatment. For completeness, γ-ratio data are also included. Since circumstances did not permit the α- and β-analysis to be carried out during the generation of the samples, the Ra^{228}/Ra^{224} activity ratios in radium-enriched samples had passed their 10–30 day maxima indicated by Fig. 9. For this reason the γ-ratio analysis was repeated on most of the samples to provide comparison data.

The α/β ratios confirm that, in the absence of γ-spectrometric facilities, this ratio serves as a rather insensitive detector of radium enrichment or depletion. The greatly reduced α/β ratios for the radium-enriched samples (C-7, M-7, M-28, M-44 and A-1) are very obvious. The Ra^{228}-enrichment of these samples is also evident in the lowering of the Th^{232}/Th^{228} ratio because of the build-up of Th^{228} in the excess of Ra^{228}. In the case of the furnace-liner samples, the purity of the radium originally in the sample is indicated by the very low Th^{232} content after 300 days as compared to the high concentration of Ra^{228}-bred Th^{228}.

EFFECT OF THORON GAS

Most of the data in these studies relate to solid samples and to radium and thorium. The behavior of thoron or Rn^{220}, the inert gas, was investigated very briefly in a series of three samples. These samples were taken at the same time on three air samplers operating at the same rate in three different locations. A control sample was taken in a non-thorium area and two samples in thorium process areas, one over a thorium heating bath and the other over a thorium machining operation. By rapid ashing and dissolution of the filters to prepare evaporate deposits on counting planchets, it was possible to begin α-spectrometry within $1\frac{3}{4}$ hr after sampling had stopped.

The first α-spectrum from each of the samples is shown in Fig. 12. The much greater abundance of the Bi^{212} and Po^{212}, the α-emitters of the Rn^{220} series, as compared to the Po^{214} daughter of the Rn^{222} series from natural radon is very obvious in the samples from the thorium-processing areas. These α-daughters of radon and thoron are collected rather than the radon and thoron themselves because the latter are inert

Table 2. Alpha-, β- and γ-analysis results on selected samples

Sample number	Description	Date of analysis (days after melting or use)	Ra^{228}/Ra^{224} activity ratio, γ-method	α/β count rate ratio	Th^{232}/Th^{228} activity ratio
C–3	Rod starting material	—	0 .98	0.79	2.23
C–7	1st melt, 1st cut, 1st ingot	27	6.2	—	—
		265	2.3	0.52	1.30
C–15	1st melt, 2nd cut, 1st ingot	32	0.4	—	—
		265	0.7	0.99	1.95
C–17	1st melt, 2nd cut, 1st ingot	32	2.6	—	—
		265	1.9	0.60	1.08
C–38	2nd melt, 1st cut, 1st ingot	14	0.65	—	—
		246	0.83	0.85	2.10
C–34	2nd melt, interior cut, 1st ingot	14	0.18	—	—
		246	0.38	0.93	2.19
C–131	2nd melt metal, final machining	48	0.15	—	—
		259	0.38	1.05	2.28
M–5	"Pickle" solution–rod starting material	68	1.6	—	—
		313	1.4	0.97	1.30
M–7	"Pickle" solution–1st melt ingots	28	21.3	—	—
		315	3.4	0.43	0.33
M–8	"Pickle" solution–2nd melt material after machining	47	0.17	—	—
		315	0.41	1.09	1.56
M–28	1st melt furnace liner smear	14	38.5	—	—
		281	4.6	0.32	0.05
M–44	2nd melt furnace liner smear	56	43.8	—	—
		252	5.3	0.39	0.06
A–1	Air sample during unloading of 2nd melt ingot from furnace	54	48.3	—	—
		309	4.4	0.41	0.13

gases which pass through the paper filter, leaving only their daughter products—metallic ions which attach to any available fiber or particle. In addition, the short half-life of thoron would eliminate any chance of measuring it directly under these circumstances.

The purpose of these brief experiments was to learn something of the thoron content of the atmosphere near thorium operations as compared to non-thorium atmospheres. The data from these experiments are summarized in Table 3 to show the relative contributions of the thoron series and to indicate the length of elapsed time required between collection and gross α-counting in order to insure that the data from an air sample are not indicating merely the presence of the relatively harmless thoron-series activity.

The data indicate that thorium-environment air samples may have 3.5 times as much short-lived activities as other samples, some 2 hr after collection. In the thorium area samples, the thoron products are a much greater contributor to this activity than in non-thorium samples, by a factor of about 9.

RADIOLOGICAL HEALTH IMPLICATIONS

Without attempting a study of the radiological hazards of thorium,[5–7] several comments can be made regarding the evident radiological health implications of the radioactivity distribution patterns observed in the processing, particularly arc melting, of thorium.

(1) Arc melting effectively removes the radium and other daughters from thorium, but all of the daughters except Ra^{228} and Ac^{228} return very quickly and remain with the Th^{228} present.

Table 3. Thoron series activity in airborne particulate samples

Sample description	Time after collection (hr)	Thoron daughter content (% of radon–thoron total)	Apparent air contamination (dis/min per m^3)
Non-thorium area	1.7	9.7	20.3
	19.3	96.2	0.9
	51.5	~100	0.3
	>300	~100	0.04
Over thorium heating bath	2.2	83.1	24.8
	20.0	95.6	7.3
	47.8	~100	2.1
	>300	~100	0.5
Over thorium machining	2.6	90.4	69.2
	21.6	94.3	23.5
	47.5	~100	5.8
	>300	~100	0.4

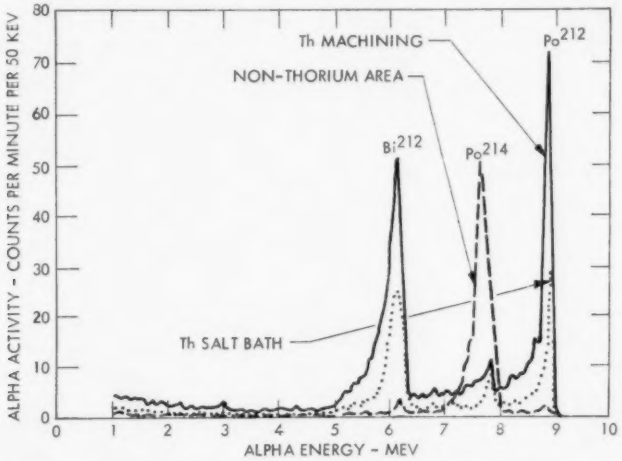

FIG. 12. α-Spectra of thorium and non-thorium-area air samples.

(2) In the arc melting of thorium of the type investigated, rather large quantities of Ra^{228}, a high-specific-activity material is released. For instance, in the casting of a 200-kg ingot of thorium such as the starting rod material, having a Th^{232}/Ra^{228} ratio of 2.3, about 9.6 mc of Ra^{228} will be removed. Probably more than one-half or at least 5 mc of this radioisotope, with a total mass of 21 μg is vaporized completely out of the ingot. The Ra^{228} is likely to be much more of a radiological hazard as 21 μg of separated material than it was distributed in 200 kg of metal.

(3) The gradual reduction of the Th^{228} and daughter activities in Th^{232} makes the detection of a given amount of thorium increasingly more difficult for methods such as *in vivo* γ-spectrometry, which are dependent upon measuring the γ-activity of the daughters.[1]

(4) These variations in the activity of the members of the thorium chain require consideration of the radioactivity pattern in each particular mass of thorium before accurate radiological health limits are imposed, as indicated by the tables of the contributions of the chain

members to the effective energy values used in calculating permissible dose.[8]

(5) The possibility of Ra^{228}, free of α-emitters, becoming airborne suggests that either simple α-counting or chemical thorium analysis, or both, is not sufficient analysis for air contamination in thorium processing.

(6) The presence of thoron daughter products requires consideration of its radiological hazard potential and also necessitates a longer waiting period before counting air samples than is usual when only the natural contributions of radon and thoron are present.

(7) In addition to the problems from thoron in air sampling, it is possible that a 3.64-day Ra^{224} could be released in the processing of thorium from which the Ra^{228} had been removed by arc melting.

CONCLUSIONS

The two important effects to be expected in thorium processing are the separation of radium and thorium and the release of thoron gas.

Enrichment or depletion of radium in samples can be best detected by observing the Ra^{228}/Ra^{224} ratio 10 days or more after the decay-chain disruption. The γ-ratio method used in these studies is useful for determining the Ra^{228}/Ra^{224} ratio because it can be applied without appreciable sample preparation, and generally to the samples in the bottles in which they are received. A satisfactory calibration for the γ-ratio method can be achieved by relating the γ-ratios on a sample of radium freshly removed from thorium to the decay of Ra^{224} in the sample.

The γ-ratio results from many samples obtained from the casting of thorium by consumable electrode arc melting indicate that the process of two arc meltings removes essentially all of the Ra^{228} from the interior of thorium ingots. Some radium is vaporized completely out of the thorium and some is distributed in the outer surface of the ingot where it can be removed by surface machining. Th-free radium is deposited on the furnace liners, where its high specific activity makes it more of a radiological health hazard than when it was distributed in large masses of thorium.

The removal of radium from thorium very gradually reduces the Th^{228} sub-series activity, making a given amount of thorium more difficult to detect by methods such as *in vivo* γ-spectrometry and causing variations in the radiological hazard of thorium materials.

The release of thoron gas in thorium processing necessitates consideration of its content in air as a radiological hazard and also as a mask of smaller amounts of long-lived activity in airborne-activity monitoring.

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BIOACCUMULATION OF RADIOACTIVE ISOTOPES BY HERBIVOROUS SMALL MAMMALS

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Abstract—This paper reports body burdens of radionuclides and dose-rate estimations for wild mammals from areas contaminated by radioactive waste disposal. Body burdens determined by radioassay are compared with body burdens estimated by an equilibrium equation. The data were gathered in a pilot-scale field study and will be used to design a more comprehensive study of large numbers of wild mammals.

The more abundant radionuclides included Sr^{90}, Ru^{106}, Cs^{137} and Co^{60}. Almost all of the mammals radioassayed had body burdens that were greater than body burdens expected from world-wide weapons fallout. The estimated whole-body dose rate from internal sources to one muskrat trapped in a settling basin for radioactive liquid waste was 112 rads/week. The estimated whole-body dose rate from internal and external sources to cotton rats trapped on the White Oak Lake bed was 2.9 rads/week. Information from a population ecology study was used with the bioaccumulation data to estimate the quantity of radionuclides held by a cotton-rat population.

INTRODUCTION

THE greatly expanded use of atomic energy in the past decade has shown that ecological information is part of the knowledge needed to evaluate the safety of existing procedures and the standards needed for future expansion of the nuclear industry. One of the immediate problems of radioecological importance involves the disposal of radioactive wastes, especially intermediate- and low-level wastes which are present in large volumes at atomic energy installations. Low-level wastes are frequently disposed of to the general environment for dilution in air and water, and for fixation in soil. A potential hazard could result when certain organisms concentrate in their tissues radionuclides from low-level wastes. The evaluation and solution of environmental hazard problems requires a detailed knowledge of the fates and biological effects of radioactive materials in the environment.

Although the radiobiology of standard laboratory animals has been extensively investigated, the radiobiology of native animals has not. Little is known about the applicability to wild mammals of health physics standards obtained by laboratory experiments. Are calculations and predictions based upon biological half-lives, assimilation factors, critical organs, etc., determined for laboratory rats valid for native rats? Often replicate laboratory experiments yield results which do not agree because conditions were not strictly duplicated. Even greater differences might be expected between laboratory and field experiments which represent two extreme conditions; laboratory strains of animals are genetically homogeneous, whereas wild animals are genetically heterogeneous. Certain radioecological and health physics predictions about wild mammals can be made from existing knowledge, but these predictions should be checked by field research. Such knowledge obtained from field research might be put to immediate use in selecting the locations and the sizes of areas being designated for radioactive waste disposal and to better manage existing waste-disposal areas. The ability to predict levels of contamination in different tissues of wild mammals or even the ability to

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

1 PROCESS WASTE TREATMENT PLANT AND SETTLING BASIN
2A SOLID WASTE BURIAL GROUND NO.1
2B " " " " NO.2
2C " " " " NO.3
2D " " " " NO.4
2E " " " " NO.5
3 SEWAGE TREATMENT PLANT EFFLUENT
4 LAUNDRY EFFLUENT
5 REACTOR COOLING WATER
6 LIQUID WASTE PITS
7 HOMOGENOUS REACTOR TEST WASTE
8 DRY BED OF FORMER WHITE OAK LAKE
BETHEL VALLEY ROAD
ORNL AREA
WHITE OAK CREEK
MELTON CREEK
CI.R MILE 20·8
WHITE OAK DAM AND MONITORING STATION
W.O. Cr. MI. 0·6
CI.R MILE 21
WHITE WING BRIDGE
CLINCH RIVER
1000 0 1000 3000 5000 7000
FEET

FIG. 1. Sources of radioactive contamination in the White Oak Creek drainage system.

predict accurately which species of mammals would be least contaminated following a nuclear attack would also be of importance, because wildlife might be utilized as an emergency source of food.

The Oak Ridge Reservation of the U.S. Atomic Energy Commission has several sources of contamination from low-level and intermediate-level wastes (Fig. 1). Almost all of the nonairborne contamination at Oak Ridge passes through the White Oak Creek drainage system, which empties by way of White Oak Creek into the Clinch River at mile 20.8. The former White Oak Lake and the dry bed which was formed after the lake was drained in 1955 have been the environs for pioneering studies in radioecology. Early work on bioaccumulation of radioisotopes was done with the fish populations and other aquatic organisms.[1,2] Since the lake was drained, work has been reported on the movement of radionuclides through the soil, plant, insect and bird compartments of the lakebed ecosystem.[3–7]

Interpretation of radioisotope bioaccumulation by small mammals in contaminated habitats requires a basic knowledge of the ecology of the populations. Since information on population ecology is now available from a three-year population investigation,[8] the next planned step was a pilot study to determine the quantities and distribution of radionuclides in the herbivorous small mammals, and to make

preliminary estimates of dose rates and radioisotope equilibrium relationships between the animals and their food. Effort was concentrated on obtaining herbivorous small mammals because the path of radionuclide uptake from the soil to these mammals is easier to trace than are the paths of uptake for animals which are more catholic in their diets or for larger mammals which range over larger areas. This pilot study was prerequisite for the third step, which will be a comprehensive study of somatic effects of radiation, dose rates, and the role of the mammals in the biogeochemical cycling of radionuclides.

MATERIALS AND METHODS

Muskrats and rabbits were caught with Conibear traps, and rats and mice were captured with Museum Special snap-traps. Both kinds of traps kill instantly, making it possible to use stomach contents to estimate the kinds and concentrations of radionuclides in the diet. Tissues from the freshly killed animals were prepared for γ-spectrometric analysis by drying them in a vacuum oven at 40 ± 0.5°C. The exclusive use of a vacuum oven permits rapid drying at low temperatures and prevents the loss of volatile materials such as the tetroxide form of Ru^{106}. A γ-spectrometer equipped with a flat 3×3-in. NaI crystal and a 200-channel pulse-height analyzer was used for all γ-scans. Radiochemical analyses for Sr^{90} were performed according to standard techniques with the determinations done on an anticoincidence β-counter.

RESULTS AND DISCUSSION

Forty-two small mammals from six locations were radioassayed in the spring and summer of 1960 and yielded over 800 quantitative identifications of radioisotopes. These data are difficult to treat in a concise manner because several different species are involved, some of the variations in bioaccumulation are large, and because each area represented a different ecological situation. In this paper, data are discussed for three of these areas (a waste treatment settling basin, surface seeps of liquid waste pits, and lower White Oak Lake bed). These areas are important from both ecological and health physics standpoints, and the combined data of the three areas are illustrative of the principal findings of the pilot study. Table 1 lists the animals from each area which are treated in the discussion.

Settling basin

During the times animals were sampled from the settling basin, and for several years previously, this million-gallon impoundment was used to allow the settling of particulates onto which radionuclides were sorbed and to provide a hold-up time for radioactive decay of short-lived radioisotopes. Recently the use of this facility

Table 1. *Species list of small mammals radioanalyzed from three areas of environmental contamination in White Oak Creek drainage System*

Area	Species radioassayed	Number of individuals
Settling basin	Muskrat (*Ondatra zibethicus*)	3
East surface seeps of liquid waste pits	White-footed mouse (*Peromyscus leucopus*)	2
	Golden mouse (*Peromyscus nuttalli*)	3
	Cotton rat (*Sigmodon hispidus*)	1
Lower White Oak lake bed	White-footed mouse (*P. leucopus*)	2
	Pine-mouse (*Microtus pinetorum*)	1
	Cotton-tail rabbit (*Sylvilagus floridanus*)	4
	Cotton rat (*S. hispidus*)	5
	Muskrat (*O. zibethicus*)	1
	Total	22

Table 2. Concentrations and body burdens of radionuclides in two muskrats from settling basin

Specimen number		Concentration of radionuclide ($\mu\mu c$/g, dry wt.)				
		Cs^{137}	Zn^{65}	Co^{60}	Ru^{106}	Sr^{90}
♀ 202SVK	Pelt	9880	487	392	*	2060
♂ 204SVK	Pelt	9140	1080	895	*	2160
♀ 202SVK	Stomach and contents	7390	*	11,800	14,300	17,700
♂ 204SVK	Stomach and contents	10,100	*	24,500	*	73,600
♀ 202SVK	Intestinal tract and contents	27,400	*	36,100	13,300	58,100
♂ 204SVK	Intestinal tract and contents	10,700	*	773,000	*	139,000
♀ 202SVK	Residual carcass	18,700	608	446	*	81,600
♂ 204SVK	Residual carcass	7710	1400	838	*	70,500
		Burden of radionuclide (μc)				
		Cs^{137}	Zn^{65}	Co^{60}	Ru^{106}	Sr^{90}
♀ 202SVK	Whole animal less GI tract and cont.	3.710	0.053	0.098	*	13.800
♂ 204SVK	Whole animal less GI tract and cont.	1.140	0.193	0.122	*	8.370
♀ 202SVK	GI tract and cont.	0.639	*	0.859	0.394	1.370
♂ 204SVK	GI tract and cont.	0.976	*	67.600	*	12.400

* None detected.

was discontinued. The basin received treated wastes from a Process Waste Treatment Plant and untreated liquid wastes which were discharged directly from certain laboratory operations. The large volume of untreated waste effluent draining from the settling basin was the principal source of radioactive contamination in White Oak Creek and the Clinch River.[9] Presently all liquid wastes go to the Waste Treatment Plant and the treated effluents are released to White Oak Creek.

Muskrats have been inhabiting the settling basin for many years. A highly radioactive muskrat from the vicinity of the settling basin was found to have an osteogenic sarcoma of the right tibiofibula.[10] It was calculated that the muskrat was receiving a dose at the rate of 40 reps/day from the radioisotopes deposited in its tissues. The skeletal burden of Sr^{90} in the affected leg was high enough to produce a clear autoradiogram.

Three muskrats from the settling basin were trapped and radioanalyzed for the pilot study Two of the animals were divided into pelt, stomach with contents, intestinal tract with contents, and residual carcass, while the third animal was divided into major organs. Results of the spectrometric and radiochemical analyses of the first two muskrats are listed in Table 2. The concentrations of individual radionuclides in the two muskrats range from about 10 to 100,000 times greater than expected average concentrations in mammals exposed only to general world-wide weapons fallout. Variations in bioaccumulation of radioisotopes by these muskrats were due in part to differences in the radioisotope composition of the liquid in the settling basin from day to day, and to the different times of capture for the specimens. Specimen 204SVK had an unusually large concentration of Co^{60} in its intestinal tract, but its stomach, residual carcass, and pelt were at a

level comparable with specimen 202SVK. It seems apparent that this large amount of radioactivity was due to a recent feeding, and part of it probably would have appeared in the organs and tissues several hours later after sufficient time for intestinal absorption and transport by the blood. Zinc-65 was not detected in the GI tracts of the two muskrats but was present from past ingestions in significant concentrations in the pelt and residual carcass.

The total amounts of radioactivity in the two muskrats (Table 2) were computed from the concentrations in Table 2 and the weight of each fraction. The pelt and residual carcass of each muskrat were combined to give the whole-body burden, less GI tract with contents. Stomach with contents and intestinal tract with contents were combined to give the total amount of radioactivity in the GI tract with contents. Specimen 204SVK has about 25 times more radioactivity in its GI tract with contents than specimen 202SVK; however, the whole-body burdens of the two animals show reasonable agreement (Table 2).

A standard dose-rate equation was used to estimate the whole-body dose rate to each muskrat from (1) radionuclides deposited in the tissues only, and (2) radionuclides in tissues and GI tract with contents. The ICRP[11] equation for dose rate, R rads/week, to an organ of m g which contains a burden of qf_2 μc is

$$R = \frac{qf_2E_i}{2.8 \times 10^{-3}\, m}. \qquad (1)$$

E_i is the effective absorbed energy in MeV per disintegration. To obtain an estimate of the average dose rate to the whole body, let $f_2 = 1$.[11]

Values for E_i used in the calculations are: Sr^{90}–Y^{90}, 1.1; Cs^{137}, 0.41; Co^{60}, 0.72; Ru^{106}–Rh^{106}, 1.4; and Zn^{65}, 0.15. These values apply to a 10-cm-dia. organism, which is the approximate diameter of a muskrat. The dry-weight mass (m) of specimen 202SVK was 254 g and of specimen 204SVK was 233 g. The body burden q for each animal was obtained from Table 2.

The estimated dose rate to the whole body from radionuclides deposited in the tissues of 202SVK is 23.7 rads/week (90.4 per cent due to Sr^{90}) and for 204SVK the dose rate is 15.1 rads/week (94.0 per cent due to Sr^{90}). Since the radionuclides in the GI tract are not included, these estimates are probably minimum values. When the radionuclides in the GI tract with contents are included, the estimated dose rate to 202SVK is 27.8 rads/week and the estimated dose rate to 204SVK is 112 rads/week. The latter estimates are probably maximum estimates because the self-absorption factor is neglected. The sevenfold increase in the dose rate for 204SVK estimated by inclusion of the GI tract with contents resulted mostly from 67.6 μc of Co^{60} and 12.4 μc of Sr^{90} in the contents of the intestinal tract.

Besides the doses from internal sources, these muskrats also received an additive external dose not yet estimated, which was due to their swimming in radioactive water and burrowing in radioactive mud and soil.

Concentrations of radionuclides on a per gram dry weight basis in major organs and tissues of the third muskrat (201SVK) were computed to determine the distribution of the radionuclides in the body and are listed in Table 3. Cesium-137 was found in all samples, and Co^{60} was found in all samples except eyes and lower incisor teeth. Cerium-144 and Ru^{106} were found only in the contents and walls of the GI tract; on the other hand, Zn^{65} was absent from the contents of the GI tract but was present in most of the tissues and organs, as was found for muskrats 202SVK and 204SVK (Table 2). Zinc-65 is the only radionuclide which was removed from the pelt by washing (Table 3). Concentrations of Sr^{90} were higher in teeth and bone than in stomach contents, indicating that this biologically dangerous radioelement is being accumulated in the tissues to concentrations exceeding those in the diet.

East seepage area of liquid waste pits

Five liquid waste pits have been dug a few hundred yards northwest of White Oak Lake bed. Four of these pits are still in service; the oldest one was experimental and is no longer used. The pits function as storage basins and as ion exchange columns for the liquid that seeps through the Conasauga Shale formation.[12,13] Almost 500,000 c of mixed radioisotopes have been disposed of to Pit Nos. 2, 3 and 4 since

Table 3. Concentrations of radionuclides in tissues and organs of muskrat ♂ 201SVK from settling basin

Sample	Concentration of radionuclide ($\mu\mu c$/g, dry wt.)						
	Cs^{137}	Cs^{134}	Co^{60}	Ce^{144}	Ru^{106}	Zn^{65}	Sr^{90}
Stomach contents	10,700	*	3620	*	5480	*	27,300
S. intestine contents	37,600	*	10,500	8010	11,000	*	44,800
Caecum and l. intestine contents	33,000	*	63,800	33,900	37,700	*	70,800
Stomach (washed)	5250	*	1160	*	*	1170	1290
S. intest. (washed)	6190	*	2090	*	786	2130	3590
Caecum and l. intest. (washed)	3990	*	8620	*	3970	*	3820
Liver	17,100	*	4240	*	*	2120	†
Kidneys	25,800	*	7560	*	*	*	†
Brain	10,600	*	285	*	*	568	†
Spleen	21,600	*	1290	*	*	*	†
Muscle	34,200	377	219	*	*	530	†
Eyes	17,300	*	*	*	*	Trace	†
Lungs	21,000	*	1080	*	*	989	†
Femurs	3280	*	160	*	*	773	190,000
Lower incisor teeth	1730	*	*	*	*	697	268,000
Pelt (washed)	5840	*	494	*	*	*	1240
Pelt (unwashed)	5540	*	406	*	*	708	1000

* None detected.
† Sample not radioanalyzed for this nuclide.

1955.[14] The external radiation levels from the γ-emitters are as high as 1 r/hr.[15] Most of the seepage occurs underground through the shale formation, but in some places the seepage reaches the surface water and forms "surface seeps" which drain across the lake bed into White Oak Creek. Monitoring operations by Waste Disposal personnel have shown Ru^{106} to be the most abundant radioisotope in the surface seepage water.

The pattern of distribution of Ru^{106} in all but one of the animals (166SVK) shows a higher concentration in the GI tract with contents than in the carcass, but the variations in concentrations are great (Tables 4 and 5). On the other hand, the variations in concentrations of Sr^{90} are small for both white-footed mice and golden mice (Table 4), suggesting an uptake mechanism which might be similar for both species. Cesium-137 in these mice is less abundant than Ru^{106}, but appears to be slightly more abundant than Zr–Nb^{95} and Co^{60}. There was a complete absence of Cs^{137} and Zr–Nb^{95} in the cotton rat.

Lower White Oak Lake bed

An arbitrary division of the lake bed into upper and lower parts has been made for ecological studies. The lower bed is not as radioactive as the upper bed, which is too radioactive in some places for personnel to work for a practical length of time. The study plot utilized for small-mammal population investigations is located on the lower bed opposite the side where the liquid-waste pits are situated. There appears to be little movement of radioactive materials from the pits across White Oak Creek, and flooding by the contaminated water from the creek covers only the lower end of the plot and parts of the plot adjacent to the creek. Radiation levels in this area are more constant than those in other collecting areas where the radiation levels often change following changes in laboratory operations. This somewhat stable situation in the lower lake bed results in less intraspecies variation of radioisotope bioaccumulation, and by combining radioisotope bioaccumulation data with population dynamics

Table 4. Concentrations of radionuclides in two white-footed mice and three golden mice from the east surface seepage area of liquid waste pits

Species	Specimen number		Concentration of radionuclide ($\mu\mu$c/g, dry wt.)				
			Ru^{106}	Cs^{137}	Zr–Nb^{95}	Co^{60}	Sr^{90}
White-footed mouse	♂ 168SVK	Pelt	838	45	40	34	†
White-footed mouse	♂ 169SVK		5030	*	*	*	†
Golden mouse	♂ 161SVK		*	37	*	*	†
Golden mouse	♀ 165SVK		*	22	*	*	†
Golden mouse	♂ 166SVK		931	*	*	*	†
White-footed mouse	♂ 168SVK	GI tract and contents	6730	*	*	795	32
White-footed mouse	♂ 169SVK		26,100	*	*	*	39
Golden mouse	♂ 161SVK		195	62	*	*	25
Golden mouse	♀ 165SVK		15,800	140	*	66	30
Golden mouse	♂ 166SVK		*	*	*	405	33
White-footed mouse	♂ 168SVK	Residual carcass	327	49	14	32	66
White-footed mouse	♂ 169SVK		1940	*	*	*	14
Golden mouse	♂ 161SVK		24	48	*	*	23
Golden mouse	♀ 165SVK		24	56	*	*	47
Golden mouse	♂ 166SVK		387	120	13	*	39

* None detected.
* Sample not radioanalyzed for this nuclide.

Table 5. Concentrations of radionuclides in cotton rat ♂ 162SVK from the east surface seepage area of liquid waste pits

Sample	Concentration of radionuclide ($\mu\mu$c/g, dry wt.)		
	Ru^{106}	Co^{60}	Sr^{90}
Pelt	3520	*	†
GI tract and contents	24,200	2260	142
Spleen	*	*	†
Kidneys	4470	190	†
Liver	4450	150	†
Femurs	970	*	1050
Residual carcass	1240	25	356

* None detected.
† Sample not radioanalyzed for this nuclide.

information it is possible to estimate the amounts of radioactivity held by a mammal population.

Animals were trapped from the edge of the lake bed where the radiation field is considerably lower than the remainder of the bed.[16] Only Cs^{137}, Co^{60} and Sr^{90} were detected in these specimens (Table 6). Most of the radioactivity was present in the cotton rat, whose range probably extended farther out on the bed than the white-footed mice or the pine mouse. The complete absence of Ru^{106} in the four animals indicates that this radionuclide may not be a contaminant in this area.

Radioanalyses of four rabbits trapped from the lake bed show that Ru^{106} is the most abundant radioisotope in the diet (Table 7). The relative abundance of Cs^{137}, Co^{60} and Zr–Nb^{95} in the rabbits is similar to the relative abundance of these radioisotopes in the white-footed mice and golden mice from the surface seeps of the waste pits (Table 4), i.e. Cs^{137} is less abundant than Ru^{106} but more abundant than Co^{60} or Zr–Nb^{95} (Table 7). Rabbits have a much larger range than small rats and mice, so that their radioisotope bioaccumulation may not be closely related to the place from where they were trapped. Strontium-90 is higher in

Table 6. Concentrations of radionuclides in one cotton rat, two white-footed mice, and one pine mouse from shoreline of lower White Oak Lake bed

Species	Specimen number		Concentration of radionuclide ($\mu\mu$c/g, dry wt.)		
			Cs^{137}	Co^{60}	Sr^{90}
Cotton rat	♂ 192SVK	Pelt	32	27	†
White-footed mouse	♂ 193SVK	Pelt	*	*	†
White-footed mouse	♂ 194SVK	Pelt	*	*	†
Pine mouse	♂ 195SVK	Pelt	*	*	†
Cotton rat	♂ 192SVK	GI tract and contents	135	64	23
White-footed mouse	♂ 193SVK	GI tract and contents	*	*	7
White footed mouse	♂ 194SVK	GI tract and contents	*	*	3
Pine mouse	♂ 195SVK	GI tract and contents	*	*	12
Cotton rat	♂ 192SVK	Residual carcass	32	60	197
White-footed mouse	♂ 193SVK	Residual carcass	8	1	95
White-footed mouse	♂ 194SVK	Residual carcass	6	*	21
Pine mouse	♂ 195SVK	Residual carcass	9	*	107

* None detected.
† Sample not radioanalyzed for this nuclide.

the femurs than it is in the stomach contents for all four of the rabbits, but is considerably higher (4.5, 7.5 and 210 times higher than stomach contents) in only three rabbits. Of all the major radionuclides in the soil, Sr^{90} is least abundant.[9]

The distribution and amounts of contamination in muskrat 137SVK (Table 8) collected from the creek in the study plot may be compared to the radionuclide bioaccumulation of muskrat 201SVK collected from the settling basin (Table 3). Most of the same organs and tissues were radioanalyzed for both muskrats. Radionuclides present in the specimen from the settling basin are about two orders of magnitude greater, which is not surprising, since the levels of contamination in the settling basin are much greater. Zirconium–Niobium-95 and Mn^{54} were detected in the muskrat from the lake bed but not in the muskrats from the settling basin. Consideration of the settling basin as the possible source of these radioisotopes has been complicated because the muskrat from the lake bed was captured four months before the capture of the muskrats from the settling basin.

Table 9 shows the whole-body concentrations and whole-body burdens of radionuclides in four lake-bed cotton rats. After substitution of the proper values into equation 1 for E_i (based upon a 5-cm-dia. organism), m and q, a whole-body dose rate of 0.3 rad/week was calculated for the radionuclides deposited in the tissues and an additional 0.1 rad/week for the contents of the GI tract. A dose rate of 2.5 rads/week was estimated for external sources by assuming total absorption of an average external radiation field of 15 mrads/hr. When the contributions from all sources are added, the total whole-body dose rate is 2.9 rads/week. The Cs^{137}, Ru^{106} and Co^{60} deposited in the lake bed soil accounts for almost all of the external radiation dose, whereas Sr^{90} accounts for 84 per cent of the dose from radionuclides in the tissues.

Partial case histories are available for three of these four rats. Rat 157SVK was first captured as an adult 80 days before the radioassay sample was taken. It is known that rats 156SVK and 158SVK were 170 days old when collected for radioassay because they were first captured, marked and released when 1 day old. It was assumed that these two rats lived the entire 170

Table 7. Concentrations of radionuclides in four rabbits from lower White Oak Lake bed

Specimen number	Sample	Concentration of radionuclide ($\mu\mu$c/g, dry wt.)				
		Ru^{106}	Cs^{137}	Zr–Nb^{95}	Co^{60}	Sr^{90}
♂ 174SVK	Stomach contents	1970	896	185	*	132
♀ 175SVK		1060	221	53	87	297
♂ 176SVK		1570	342	Trace	149	45
♀ 177SVK		54	18	*	*	6
♂ 174SVK	Feces	12,500	1070	222	*	†
♀ 175SVK		5440	862	190	505	†
♂ 176SVK		3420	894	120	407	476
♀ 177SVK		1720	140	*	*	†
♂ 174SVK	Kidneys	270	94	*	14	†
♀ 175SVK		100	67	*	32	†
♂ 176SVK		230	33	*	45	†
♀ 177SVK		170	36	*	*	†
♂ 174SVK	Liver	134	49	*	20	†
♀ 175SVK		83	44	*	21	†
♂ 176SVK		223	34	*	43	†
♀ 177SVK		58	24	*	11	†
♂ 174SVK	Muscle	*	66	*	4	†
♀ 175SVK		*	55	*	*	†
♂ 176SVK		38	38	*	5	†
♀ 177SVK		*	44	*	*	†
♂ 174SVK	Femurs	*	*	*	*	146
♀ 175SVK		24	10	*	*	1330
♂ 176SVK		25	6	*	5	327
♀ 177SVK		28	8	*	*	1260

* None detected.
† Sample not radioanalyzed for this nuclide.

days on the lake bed. Calculations were made to determine the theoretical body-burdens after 170 days and the theoretical body-burdens at equilibrium for the two rats. These estimates may be evaluated by comparing them to the observed values from radioassay. An equation which describes body burden at equilibrium is

$$Q_e = \frac{ra}{\lambda} \qquad (2)$$

where Q_e = body burden at equilibrium (μc), r = rate of feeding (μc/day), a = fraction of ingested isotope assimilated by organs, $\lambda = 0.693/T = 0.693/T_r + 0.693/T_b$, T = effective half-life (days), T_r = radiological half-life (days) and T_b = biological half-life (days). For the radioisotopes under consideration, $T \simeq T_b$ because $T_r \gg T_b$. The body burden at 170 days may be represented by

$$Q_{170} = Q_e(1 - e^{-\lambda t}) \qquad (3)$$

where Q_{170} = theoretical body burden at 170 days (μc) and t = age of animal (days). The underlying basis for use of equations (2) and (3) is that there must be a constant rate of intake of radioactivity. For the animals under discussion,

Table 8. Concentrations of radionuclides in muskrat ♀ 137SVK from creek in lower White Oak Lake bed

Sample	Concentration of radionuclide ($\mu\mu$c/g, dry wt.)							
	Cs^{137}	Co^{60}	Ce^{144}	Ru^{106}	Zn^{65}	Zr–Nb^{95}	Mn^{54}	Sr^{90}
Stomach contents	1330	130	*	1970	*	100	*	712
S. intestine contents	2440	276	1200	2400	*	120	*	494
Caecum and l. intestine contents	4220	413	3240	5050	*	280	*	941
Stomach (washed)	150	42	*	420	*	*	*	†
S. intestine (washed)	70	54	*	826	*	*	*	†
Caecum and l. intestine (washed)	59	261	*	1520	*	*	*	†
Liver	22	103	*	294	*	*	*	†
Kidneys	330	180	*	4020	*	*	*	35
Spleen	*	*	*	*	*	*	*	†
Muscle	404	*	*	*	Trace	*	*	†
Lungs	278	16	*	329	*	*	*	†
Femurs	296	*	*	*	89	*	*	9430
Lower incisor teeth	*	*	*	280	*	*	*	4710
Hair (washed)	*	*	*	330	220	*	7	66

* None detected.
† Sample not radioanalyzed for this nuclide.

Table 9. Concentrations and whole-body burdens (exclusive of GI-tract contents) of radionuclides in four cotton rats from lower White Oak Lake bed

Specimen no.	Concentration of radionuclide ($\mu\mu$c/g, dry wt.)			
	Sr^{90}	Ru^{106}	Cs^{137}	Co^{60}
♀ 155SVK	648	74.1	67.9	30.9
♂ 156SVK	640	65.2	54.7	30.9
♂ 157SVK	638	36.7	47.2	18.1
♀ 158SVK	573	73.6	60.7	36.4
	Whole-body burden ($\mu\mu$c)			
	Sr^{90}	Ru^{106}	Cs^{137}	Co^{60}
♀ 155SVK	9440	1080	989	451
♂ 156SVK	12,800	1300	1090	616
♂ 157SVK	16,200	932	1200	460
♀ 158SVK	9420	1210	998	598

this is assumed. The estimated average daily rate of food intake from 0 to 170 days (6.5 g, dry wt./day) was multiplied by the average radioisotope concentration per gram of stomach contents to obtain r of equation (2). Assimilation factors and biological half-lives from the literature are:

Isotope	Assimilation factor	Biological half-life (days)
Sr^{90}	0.15[17]	350[17]
Cs^{137}	0.80[18]	6.5[18]
Ru^{106}	0.03[19]	85[19]
Co^{60}	0.30[11]	9.5[20]

The laboratory rat was used as the experimental animal for Sr^{90}, Cs^{137} and Ru^{106}, whereas the laboratory mouse was used for Co^{60}.

The Q_e and Q_{170} values are equal for both rats in terms of Cs^{137} and Co^{60}, indicating that the radioisotope concentrations in the animals theoretically were at equilibrium with the radioisotope concentrations in the food at the time of capture (Table 10). For practical purposes, equilibrium may be assumed to be reached after five biological half-lives ($\frac{31}{32}$ of the final value)

which would be 33 days for Cs^{137} and 48 days for Co^{60}. On the other hand, the estimated values in Table 10 show that the cotton rats at 170 days were slightly more than $\frac{1}{4}$ of their equilibrium body burdens for Sr^{90} and at $\frac{3}{4}$ of their equilibrium body burdens for Ru^{106}. These differences between estimates of Q_e and Q_{170} are obviously influenced a great deal by the longer biological half-lives. Because of these long biological half-lives, and the fact that cotton rats on the lake bed rarely live more than 1 year,[8] Sr^{90} concentrations in cotton rats could not come into equilibrium with concentrations in food, whereas Ru^{106} equilibrium would occasionally be possible.

Table 10. *Calculated and radioassay body burdens of radionuclides for two cotton rats from lower White Oak Lake bed*

Isotope	Body burdens ($\mu\mu c$)			
	Calculated		Radioassay	
	Q_e	Q_{170}	♂ 156SVK	♀ 158SVK
Sr^{90}	57,400	16,400	12,800	9420
Ru^{106}	11,400	8520	1300	1210
Co^{60}	9530	~9530	616	598
Cs^{137}	28,600	~28,600	1090	998

The body burden of each radioisotope determined by radioassay is in all cases lower than the calculated value, Q_{170} (Table 10). The closest agreement was for Sr^{90} where the radioassay body burdens were about $\frac{3}{4}$ of and $\frac{1}{2}$ of the Q_{170} body burden. The much larger differences for the other radionuclides indicate that either the experimental data from the literature do not apply or that the feeding rate and exposure-time estimates have significant errors. It seems likely that the most significant error might be in the time of exposure t. There is an increasing divergence between the radioassay values and Q_{170} values as biological half-life decreases. This condition would be expected if the animals had fed for a period of the time on noncontaminated food before they were captured. If $t < 170$ days, then there has not been a constant rate of intake, and use of equations (2) and (3) may give inaccurate estimates. Certainly it is possible that these cotton rats could have lived part of their lives away from the contaminated lake bed.

Estimates were made for the burdens of radionuclides present in the cotton rat populations of the lake-bed study area during population censuses in 1958, 1959 and 1960 (Table 11). The number of cotton rats per acre was calculated from trapping records by use of the population estimation formula proposed by HAYNE[21]. The average cotton-rat weight was obtained directly from the animals captured during the census, and biomass was calculated from population density and weights, with an appropriate reduction to dry weight. For estimating the concentrations of radionuclides per unit area during the times shown, a simplifying assumption was made that the concentrations per gram during these times were approximately equal to the average of the concentrations for the four cotton rats in Table 9.

The influence of population parameters on bioaccumulation of radionuclides is apparent after inspection of these data. The number of individuals per acre during the April–May 1959 census was exactly the same as during the December–February 1960 census, 8.5 cotton rats per acre; however, there is a considerable difference in the concentrations of radionuclides per acre. The reason for this difference can be found in the average individual weight, which was 100 g in one case and 120 g in the other. The reverse situation may also occur where the average individual weights are the same but the number of individuals per acre differ. This is exemplified by comparing the results of the April 1958 census with the results of the April–May 1959 census. The effect on the estimation of radionuclides per unit area is apparent. These data demonstrate how the amount of radioactivity held in this part of the mammal compartment varies depending upon the population parameters.

The cotton-rat populations on the lake bed and the noncontaminated study areas of the Reservation were exterminated after the record snow and cold weather of the 1959–1960 winter,[22] so that there were no animals for the April and July 1960 census periods. The total amount of radioactivity held by the cotton rats was given

Table 11. Cotton-rat biomass and bioaccumulation of radionuclides for lower White Oak Lake bed

Year	Month	Cotton rats per acre	Average cotton rat wt. (g)	Dry wt. biomass per acre (g)	Estimated concentrations of radionuclides per unit area ($\mu\mu$c/acre)			
					Sr^{90}	Ru^{106}	Cs^{137}	Co^{60}
1958	April	5.4	120.4	197	123,000	12,300	11,300	5730
	July	5.3	124.7	197	123,000	12,300	11,300	5730
	October, November	4.5	92.2	139	86,900	8670	8010	4040
1959	April, May	8.5	120.0	340	213,000	21,200	19,600	9890
	July, August	5.8	127.2	218	136,000	13,600	12,600	6340
	September, October, November	9.0	109.8	304	190,000	19,000	17,500	8850
1960	December, January, February	8.5	100.7	256	160,000	16,000	14,700	7450
	April	0						
	July	0						

up to the environment for recycling in the lakebed ecosystem.

CONCLUSIONS

The data obtained from radioassaying a small number of mammals from each of the six contaminated areas have served well in supplying the baseline information for designing a comprehensive mammal radioecological study. This proposed study would involve many more measurements and permit statistical treatment. The data also indicate that if a single radionuclide were to be chosen for study in detail, a logical choice would be Sr^{90}. This choice is based upon (1) the opportunity to study the physical and biological mechanisms whereby Sr^{90} as the least abundant radioisotope in the soil becomes the most abundant in the mammals, (2) the high quantity of Sr^{90} in the bones of the mammals and the good counting statistics that can be expected from these high levels, and (3) the pathological importance of chronic internal deposition of Sr^{90}. Ruthenium-106, Cs^{137} and Co^{60} should be included with Sr^{90} in a study that would involve more than one radionuclide because these radioisotopes are present in almost all the mammals. Other radioelements usually are not at high enough levels of activity to supply adequate information for reliable interpretation. Bone is obviously the critical organ for Sr^{90}, but individual critical organs for Ru^{106}, Cs^{137} and Co^{60} are not obvious from the data. The GI tract may be the best critical-organ choice for these radioisotopes. Under the conditions of chronic ingestion, the GI tract is irradiated continuously by the radioisotopes in the food, which are usually present in higher concentrations than the radioisotopes deposited in the tissues.

The lower White Oak Lake bed is the most favorable location for a more detailed study of radioisotope bioaccumulation. Relatively low intraspecies variation of radioisotope concentrations in tissues and foods indicate that contamination in this area does not change with current laboratory operations. The area is large, and the data available from a three-year study of small-mammal population dynamics on the lake bed further justifies the choice.

Biological half-lives and assimilation factors should be experimentally determined in the field for comparison with values which have been determined in the laboratory on standard experimental animals. There is an obvious need for fenced-in areas into which animals could be released and recaptured at will so that exposure time would be known exactly and thus permit more accurate equilibrium and dose-rate calculations. Where dose rates have been calculated, they have been high enough to encourage investigating for somatic effects of radiation in the mammals.

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COMPARISON OF PREDICTED AND MEASURED EQUILIBRIUM LEVELS FOR CHRONICALLY ADMINISTERED Cs^{137}*

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Abstract—The biological retention of a single oral dose of Cs^{137} was obtained by whole-body counting twelve 89-day-old female RF mice for 32 days. An IBM 704 computer curve-fitting procedure was then used to reduce the retention data into three exponential components, and the resultant retention function was integrated from t_0 to t_n to predict the Cs^{137} retention by mice of the same age at any time t_n during a chronic exposure. Both the predicted build-up of body Cs^{137} activity and the equilibrium level agreed reasonably well with measured values obtained by whole-body counting 89-day-old animals whose drinking water contained 0.012 μc Cs^{137}/ml for a 107-day period. The urinary-to-fecal excretion ratio during the period of observed equilibrium was about 9. These findings provide additional experimental verification for the current practice of using retention parameters obtained from acute experiments to predict equilibrium levels which would be attained during chronic exposure conditions.

Retention of Cs^{137} after the termination of chronic exposure did not agree with the retention pattern predicted from the original single-dose experiment, but did agree with the retention pattern predicted from a second single-dose experiment in which animals of the same age as those at the termination of the chronic ingestion period were used.

The rather obscure relation between age and retention, and consequently between age and equilibrium level or radiation dose, is discussed.

INTRODUCTION

In practice, maximum permissible concentration values are almost invariably based on data obtained from the *acute* administration of radionuclides to animals. Conceptually, however, conditions of chronic exposure are inherent to the calculations. The practice of using retention parameters obtained from acute studies to predict equilibrium levels attained during chronic exposure has rarely been subjected to experimental verification. The experiments of Cook *et al.*[1] using Co^{60} in mice, and those of Ballou and Thompson[2] using Cs^{137} in rats, probably represent the most rigorous attempts to test the validity of this practice. These studies are acknowledged in the most recent Recommendations of the International Commission on Radiological Protection[3] as providing fairly satisfactory evidence for extrapolating from acute dose studies to the situation corresponding to chronic exposure. All the evidence has not been favorable, however, as other workers,[4] using Cs^{137} in rats, reported a greater retention following chronic feeding as compared with a single administration. This report provides additional verification for the current practice of the ICRP relevant to this problem.

METHODS AND MATERIALS

Thirty 89-day-old female mice of the RF strain were used in this study. Throughout an acclimatization period and the experimental period, the mice were housed in pairs in plastic metabolism cages and allowed free access to Purina Lab Chow and water. At the beginning of the experimental period, the body weights

* Work performed under the auspices of the U.S. Atomic Energy Commission.

averaged 20.8 g. One-tenth ml of solution, adjusted to pH 5, containing 0.45 μc of carrier-free Cs^{137} as the chloride, was administered by gavage to each of twelve mice. At 30 min following intubation, the animals were counted in a 4π liquid scintillation detector. A schematic representation of this detector and the associated electronics is shown in Fig. 1. Details of the construction and performance of this counting system have been reported in detail.[5] Twenty-one subsequent measurements were made over a 32-day period to determine the retention of Cs^{137} as a function of time. At the end of the 32-day period, the initial activity had decreased by a factor of about 10^3.

Preliminary graphic analysis of the retention data suggested the presence of three exponential rates of loss. The original data were coded for analysis by an IBM 704 computer which was programmed to perform iterative least-square calculations. Coding procedures called for retention values (Y) at each time (t) and the estimated intercept and rate parameters of the retention function. Each value of Y was weighted statistically by a factor proportional to $1/Y$. The 704 procedure avoids the subjectivity associated with eye-fitting a line to a composite exponential function and, by iteration, assures that best-fit criteria are equally applicable to all components simultaneously. Integration of the resultant three-exponential component biological retention function from t_0 to t_n was then used to predict the radiocesium retained by the animals at any time t_n during chronic exposure.

Simultaneously with the acute administration, eighteen animals of the same age (89 days) were allowed to drink continuously water containing 0.012 μc Cs^{137}/ml and were counted periodically over a period of approximately 100 days to measure the whole-body retention during conditions of chronic exposure. Measurements of body weight and fluid intake were made at intervals. Average daily Cs^{137} intake was calculated from the mean daily fluid intake and the Cs^{137} concentration. Urinary and fecal excretion was measured periodically throughout the chronic exposure. Checks were made for several days prior to the cessation of chronic exposure to ensure that daily intake and output of Cs^{137} were comparable during the apparent equilibrium period. At the termination of the

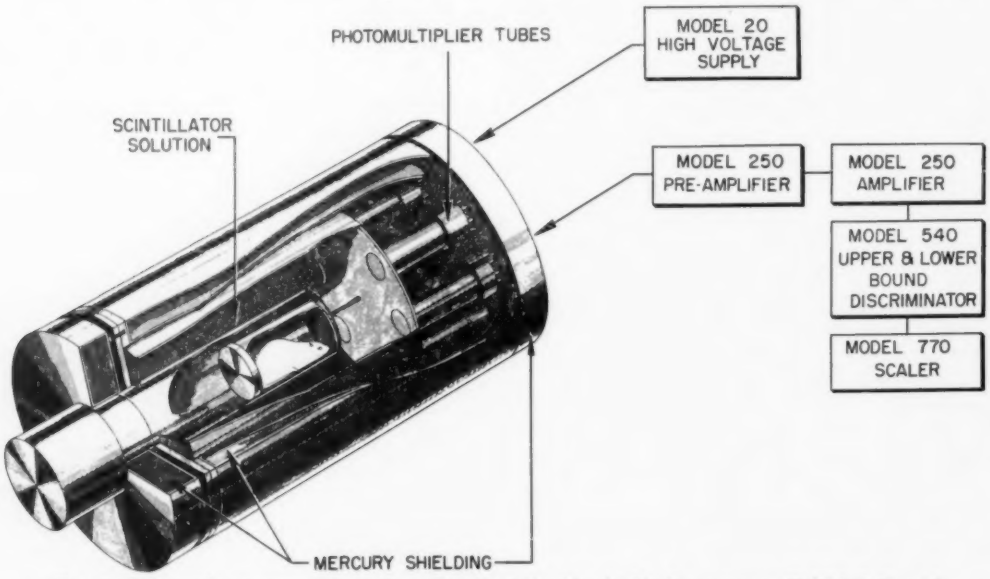

FIG. 1. Schematic representation of small-animal liquid scintillation counter (LASAC–I) and associated electronics.

chronic exposure, tap water was substituted for the Cs^{137}-contaminated drinking water, and the fall-off in radiocesium activity was studied by whole-body counting the animals.

The retention parameters obtained from the single-dose study should also predict the Cs^{137} retention after cessation of chronic exposure. However, because the animals were chronically exposed for about 15 weeks and were 15 weeks older, the acute control group was given a second oral dose of Cs^{137} coincident with cessation of the chronic experiment and was measured by whole-body counting for approximately 37 days. No residual activity from the first dose remained in these control animals at the time of the second dose. A comparison of the retention parameters could then be made between animals of ages comparable to those of the animals at the beginning and termination of the chronic exposure. This precautionary step was taken because previous experience[6] at this Laboratory strongly suggested that retention of Cs^{137} by mice varied as some function of age.

RESULTS

Retention of Cs^{137} by 89-day-old mice following a single oral exposure is shown in Fig. 2. Each point represents the mean retention value for the group of twelve mice. The iterative least-square computer analysis reduced the data into a three-component exponential function characterized by the following parameters

$$R_t = 39.09\, e^{-1.5260t} + 43.27\, e^{-0.3360t} + 17.69\, e^{-0.1474t} \quad (1)$$

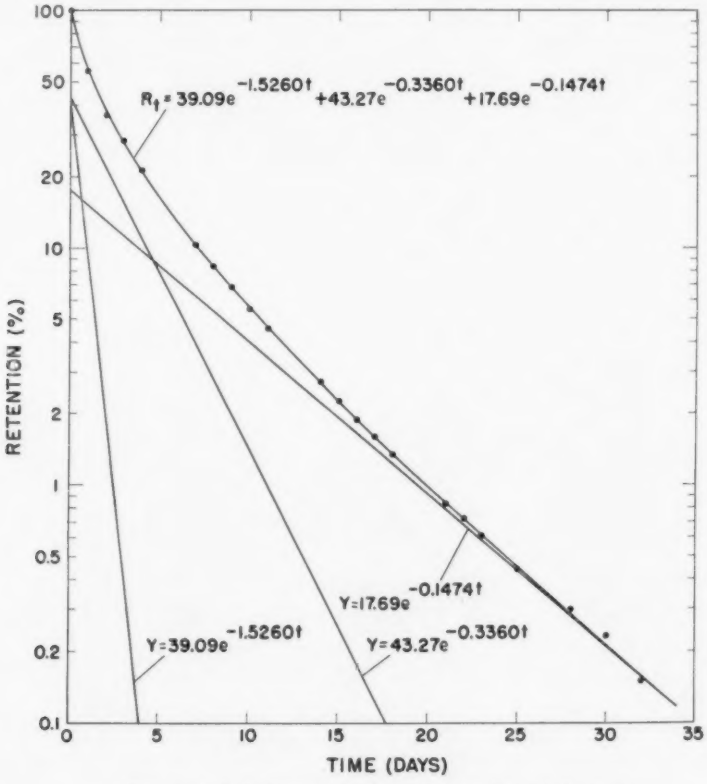

FIG. 2. Whole-body retention of a single oral dose of Cs^{137} by 89-day-old mice.

where R_t is the whole-body retention at any day t, e is the base of natural logarithms, and the coefficient and exponent of each term are the intercept and rate constant. The line drawn through the experimental points was calculated from the retention function and gives an indication of the precision of the mathematical description of the data. Also shown in Fig. 2 are the three exponential components which contribute to the composite retention function.

Fig. 3 shows the predicted retention pattern and the values measured for eighteen mice during the chronic exposure period. The left-hand ordinate indicates the build-up to equilibrium for each component and for the sum of the three components. The right-hand ordinate equates the equilibrium level to 100 per cent retention. Although the first component represents about 39 per cent of the single oral dose, it accounts for about only 10 per cent of the total body burden when equilibrium conditions are established, because of the very short half-life (0.45 day).

Fig. 4 shows both retention and excretion of Cs^{137} by the mice during the chronic exposure period. The total excretion data indicate that daily intake and output of Cs^{137} are roughly equal from about 21 days to the end of the experimental period. As approximately 10 per cent of the daily Cs^{137} intake is lost by fecal excretion, the urinary-to-fecal ratio is about 9.

Based on the parameters obtained from the single administration experiment, the predicted fall-off in retention following cessation of chronic exposure should be

$$R_t = a_1/k_1\, e^{-k_1 t} + a_2/k_2\, e^{-k_2 t} + a_3/k_3\, e^{-k_3 t} \quad (2)$$

or, in terms of per cent of the equilibrium level, should be

$$R_t = 9.33\, e^{-1.5260t} + 46.93\, e^{-0.3360t} + 43.74\, e^{-0.1474t} \quad (3)$$

The retention curve obtained from the solution of equation 3 is shown as a broken line in Fig. 5. The measured retention data following cessation of chronic administration, as shown in Fig. 5, are a poor fit to the line predicted by equation 3. The solid line, which is a reasonably good fit to the retention data, was calculated from the parameters which describe retention following a single oral administration to the control animals coincident with termination of the chronic exposure period. Thus, both groups of animals were the same age at the cessation of chronic exposure. The solid line was calculated from the following retention function

$$R_t = 42.56\, e^{-1.60946t} + 20.92\, e^{-0.21962t} + 36.41\, e^{-0.11474t} \quad (4)$$

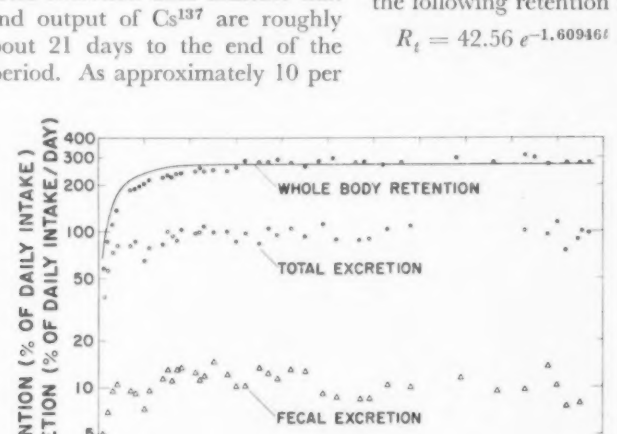

FIG. 3. Comparison of predicted and measured whole-body retention of chronically administered Cs^{137} by 89-day-old mice.

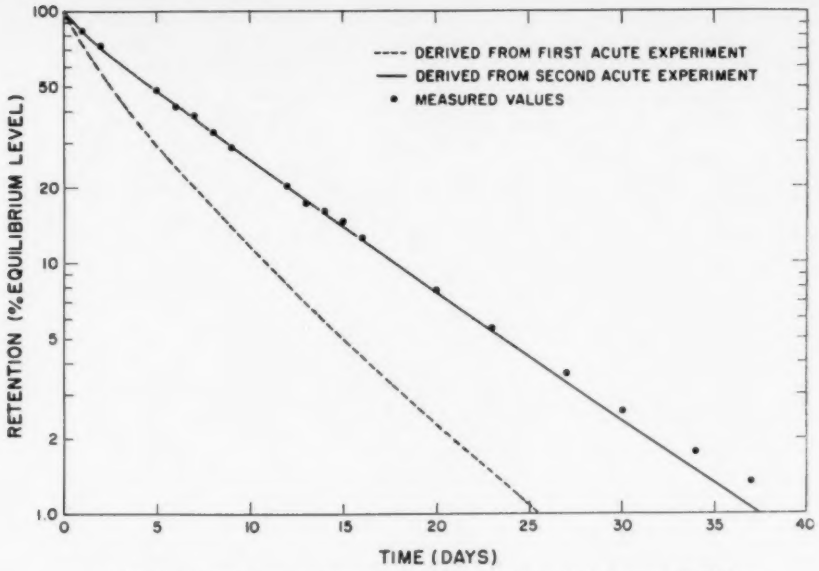

FIG. 4. Retention and excretion of chronically administered Cs^{137} by 89-day-old mice.

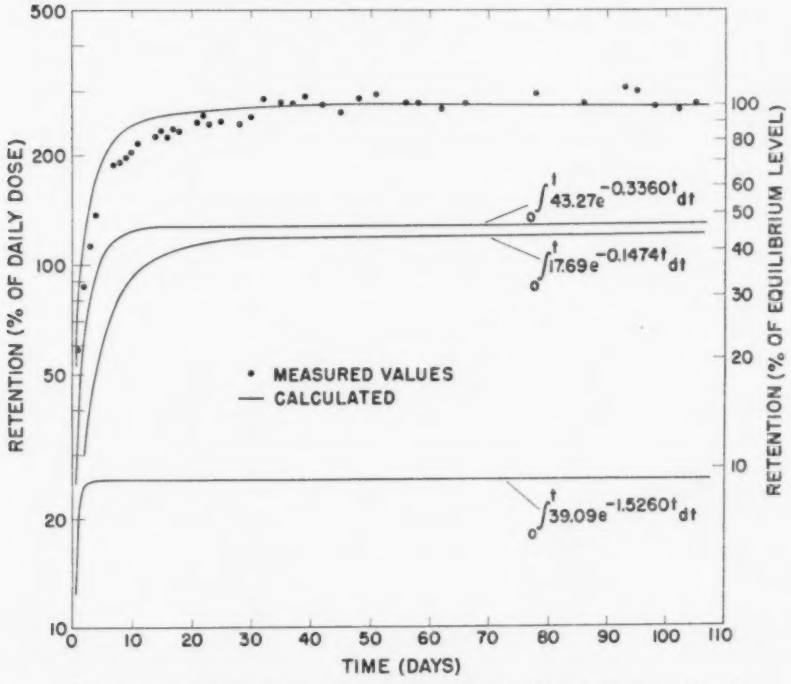

FIG. 5. Comparison of predicted and measured whole-body retention of Cs^{137} by 195-day-old mice following cessation of chronic exposure.

DISCUSSION

Although the approach to equilibrium in body Cs^{137} activity during chronic ingestion does not precisely follow that predicted from a single-dose study, the data presented here indicate that the actual equilibrium level attained during chronic ingestion may be predicted with considerable confidence. However, failure of the animals to lose Cs^{137} according to the pattern predicted from the acute retention parameters, except when animals of the same age are used, implicates age as a factor which complicates the prediction of retention following cessation of chronic exposure—at least for animals of short life span. Of considerable interest is the finding that the measured and predicted equilibrium levels agree quite well, even though the chronically exposed animals were measured for about 15 weeks. One might reasonably expect the measured equilibrium level to change with time during exposure as the animals are continually aging. This phase of the problem will be investigated by studying both the long-term retention of chronically administered Cs^{137} and the retention of single doses of Cs^{137} administered at intervals throughout the life span of the mouse.

It is important to stress here that the equilibrium levels predicted from studies on animals given a single dose (Figs. 2 and 3) should be applied only to animals of comparable age, as retention of a single oral dose of Cs^{137} appears to vary as some function of age. This restriction is not important at present for human subjects as, in general, little information is available for most radionuclides regarding metabolism by man. We have shown[6] that for mice given Cs^{137} intraperitoneally, the rate constant of the slowest changing component decreases by about 20 per cent between 4- and 20-week-old animals. Rate constants for 20-, 32-, 51-, 74- and 88-week-old animals remained relatively constant at about 0.1245 day^{-1}. More striking were the values for the area under the retention functions integrated from t_0 to t_∞, which varied with age as shown in Table 1. These areas are proportional to both radiation dose and equilibrium level. Similar results were obtained by us for juvenile and adult rats given a single injection of Na^{22}.[7] Younger animals lose Na^{22} more rapidly initially but incorporate more Na^{22} in the slowest changing component than older animals.

Table 1. Total area under biological retention functions for Cs^{137} given intraperitoneally to mice of different ages*

Group	Age at injection (weeks)	Area (% days)
A	4	386
B	12	379
C	20	423
D	32	460
E	51	500
F	74	468
G	88	470

* Area under retention function integrated from t_0 to t_∞, i.e. the total area is $\sum_i^n \left[\frac{a_i}{k_i}\right]$, when retention function is $\sum_i^n [a_i e^{-k_i t}]$. Because of the long half-life, loss due to physical decay of the Cs^{137} can be ignored.

The exact relation between age and retention is not obvious from reports which appear in the literature. Van Dilla *et al.*[8] observed a decrease in radium retention by beagles as the age at injection increased. Glad *et al.*[9] observed a similar age effect for beagles given Sr^{85}. Except for juvenile animals, however, the major differences among the retention functions (power functions) for the different age groups were in the coefficient rather than in the exponent. This implies that the uptake by the compartment, rather than the turnover by the compartment, is somehow age-dependent. Age effects were also observed in rats given radiostrontium,[10,11] and in cattle injected with Ca^{45}.[12] Hood and Comar[13] observed age-dependent changes in the retention of Cs^{137} by rats. Older animals retained Cs^{137} more tenaciously than younger ones. It would appear that materials which localize in the soft tissues tend to be lost at faster rates in younger animals than in older ones, possibly as the result of differences in metabolic rates. The picture is not as simple for materials which are not quantitatively absorbed from the gastrointestinal tract and which subsequently localize in hard tissues. It would appear that age-dependent

gut-absorption factors contribute strongly to and, therefore, complicate the over-all dependence of retention on age.

Studies similar to those reported here will be undertaken for materials which localize primarily in hard tissues and also for materials whose retention can be described either by the sum of exponentials or by a power function.

Acknowledgment—The authors sincerely acknowledge the valuable counsel and assistance of W. LeStourgeon (Group T–1) in deriving the parameters of the retention functions and the equilibrium curve.

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NOTES

Survival of Waste Containers at Ocean Depths

(*Received* 26 *July* 1961)

For over ten years the National Institutes of Health, with the co-operation of the United States Coast Guard, have been disposing of radioactive wastes in the Atlantic Ocean.[1] All disposals have been made at depths exceeding 6000 ft, in an area about 80 miles due East from the coast at Norfolk, Virginia. Wastes have been packaged in either 55-gal steel drums or in reinforced-concrete burial vaults, children's size. The vaults are somewhat more expensive but are easier to process, ship and put overside.

Wastes are prepared for disposal by combining them with a thin concrete mix, added in small increments during vigorous stirring. All plastic bags and other containers of solids are ruptured during the mixing. Liquids are placed in various types of jars, bottles or cans, each being filled as completely as possible. Voids in the finished product are of course undesirable, because they reduce the over-all density and lower the resistance to crushing by hydrostatic pressure. The shallow, open-top vaults have definite advantages over the taller drums during mixing operations. Both types of containers are finished with about 4 in. of a stiff concrete mix with no added wastes.

The tests to be described were undertaken to determine the effects of high hydrostatic pressure on the two types of containers. Two vaults and two drums were prepared in the usual way except that no radioactive materials were included. No undue care was taken in the preparation of the test containers. Representative laboratory wastes included animal-cage cleanings, paper, sample bottles, and counting planchets. Concentrated solutions of a black dye were put in 1-gal paint cans, and earthenware crocks.

Each finished vault was placed inside a loose-fitting cage of expanded metal which permitted the free entrance of sea water but which would hold all but small pieces in case of extensive disintegration under pressure. Each finished vault weighed about 1500 lb in air, each drum about 800 lb. Corresponding weights in water were about 750 and 450 lb.

For each test, one vault and one drum were shackled about 45 ft apart on a 130-ft length of $1\frac{1}{8}$-in. chain. The free end of this chain was shackled to an 8000-ft length of $\frac{5}{8}$-in. dia. nylon rope. This rope, without splices, had a nominal breaking strength of 10,000 lb. The line was handled by one of the main towing drums of the United States Coast Guard Cutter, *Cherokee*.

Test lowerings were made 15 days after the containers were prepared, in the vicinity of Lat. 36°57′N, Lo. 74°25′W at a depth of 1025 fathoms. In the first test, with the drum running free, the load reached the bottom in about 25 min. Recovery started within an hour and required 2 hr, 45 min. Fig. 1 shows the first test pair after recovery.

In the second test, the free end of the nylon line was shackled to 150 ft of 1-in. chain which was in turn shackled to a lighted buoy. The entire rig was put overside and left for 26 days when recovery was made as in the first test. The upper length of chain kept the vulnerable nylon line below depths at which it might be damaged by sharks.

Each of the four containers showed mud on all surfaces, indicating that it had indeed been on the bottom. Evidence for the depth of penetration into bottom mud is inconclusive. During the first test the ship was held at anchor by the two containers in the presence of a light breeze, and a surface sea current estimated to be about 2–3 knots. It seems unlikely that this would be possible with a 1200-lb load unless there had been some burial. A positive statement cannot be made on this point because of the lack of information on anchoring at these depths. Dynamometer readings taken during the second recovery showed no sudden drop as might be expected if a large object suddenly broke out of mud. However, this observation is also inconclusive for a sudden drop in tension might be masked by the remarkable elastic properties of nylon, damped by 6000 ft of water.

None of the recovered containers showed signs of gross injury. The steel drums showed some depressions at places where compressible waste materials had not been intimately mixed with concrete (Fig. 2). There was no cracking or other failure of the vaults.

The lack of fractures in the relatively thin walls of the vaults shows that water entered promptly between the walls and the contents. This is to be expected since the concrete filling was not bonded

pressure-tight to the walls. The deformations in the steel drums must have resulted from an initial, high pressure differential, but the condition of the contents indicated that water eventually penetrated throughout the interior.

Detailed examinations were made by stripping off the outer shells and carefully chipping away the concrete to expose the contents. The most surprising feature was the poor mixing of concrete and waste material. Large volumes of paper, in particular, showed no contact with concrete, in spite of what had seemed to be vigorous and thorough mixing during preparation.

Several liters of free sea water came out of each container during dissection and all of the solid wastes were water soaked. All of the concrete surfaces fractured during dissection were moist, indicating that the high pressure had forced sea water through the body of the concrete itself. This suggestion is borne out by the appearance of freshly fractured surfaces of the earthenware crocks. One of these crocks had fractured early in the test, while the other was recovered intact, still filled with dye solution. Test fractures showed dye stains part way through the walls, as if external pressure had forced liquid into the ceramic material.

All paint cans were recovered with contents intact. All were wrinkled by external pressure because of slightly incomplete filling, Fig. 3. Incompletely filled bottles had their stoppers forced in or ruptured.

Conclusions

These tests demonstrate that concrete-filled containers remain essentially intact under the high hydrostatic pressures at ocean depths. This is due to the lack of a solid bond between the container and the filling. Since sea water penetrates throughout the structure, all the contents are to some degree in contact with surrounding sea water. Most of the contacts will be tenuous, so that transfer to surrounding water by diffusion should be slow. Liquid wastes can be completely isolated by containers which yield without failure until pressures are equalized. Metal containers resistant to corrosion should provide isolation for long periods of time.

The mixing of waste products and concrete was disappointing, being much less complete than we had supposed. This mixing can undoubtedly be improved and can be studied without taking the containers to depths.

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Radiation Safety Office J. F. Thompson*
National Institutes of Health H. L. Andrews

* Commander, United States Coast Guard.

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Tests Carried Out on Organic Cation Exchange Resins for the Decontamination of Aqueous Radioactive Effluents

(*Received* 22 *September* 1961; *in revised form* 24 *October* 1961)

We have considered using resins in their sodium form for decontamination, to achieve concentration below the permitted limits, of the effluents released by the laboratories and workshops of the Centre d'Etudes Nucléaires at Saclay, and whose level of radio-activity due to anions is negligible. The resins are in fixed-bed columns and are regenerated by NaCl solution. The activity is recovered in a volume of eluent which is on the average twenty times smaller than that of the radioactive waste. This smaller volume is efficiently decontaminated and purged of its ions other than sodium by a two-stage chemical treatment which gives, for a given decontamination factor (DF), a concentration factor (CF)* higher than that of the most favourable coprecipitation treatment which can be effected on the whole of the active residues. The regenerator thus treated can be used for several successive elutions before being disposed of after decontamination. We give below the results obtained on an actual effluent which was used for our test experiment series in the laboratory:

Characteristics of the effluent:
- Amount of salt in the residual water after a filtration to give a clear liquid = 2·8 g/l.
- Total activity : $1{\cdot}3.10^{-2}$ c/cm³
- ^{90}Sr–^{90}Y activity : $5{\cdot}4.10^{-3}$ c/cm³
- ^{137}Cs activity : $3{\cdot}8.10^{-3}$ c/cm³

Results of a cycle of five fixations and regenerations on resins:
- DF total : 100
- CF : 125

Results of a double chemical treatment by coprecipitation:
- DF total : 75
- CF : 50

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*Concentration factor = $\frac{\text{effluent volume treated}}{\text{active residue volume to be conserved}}$

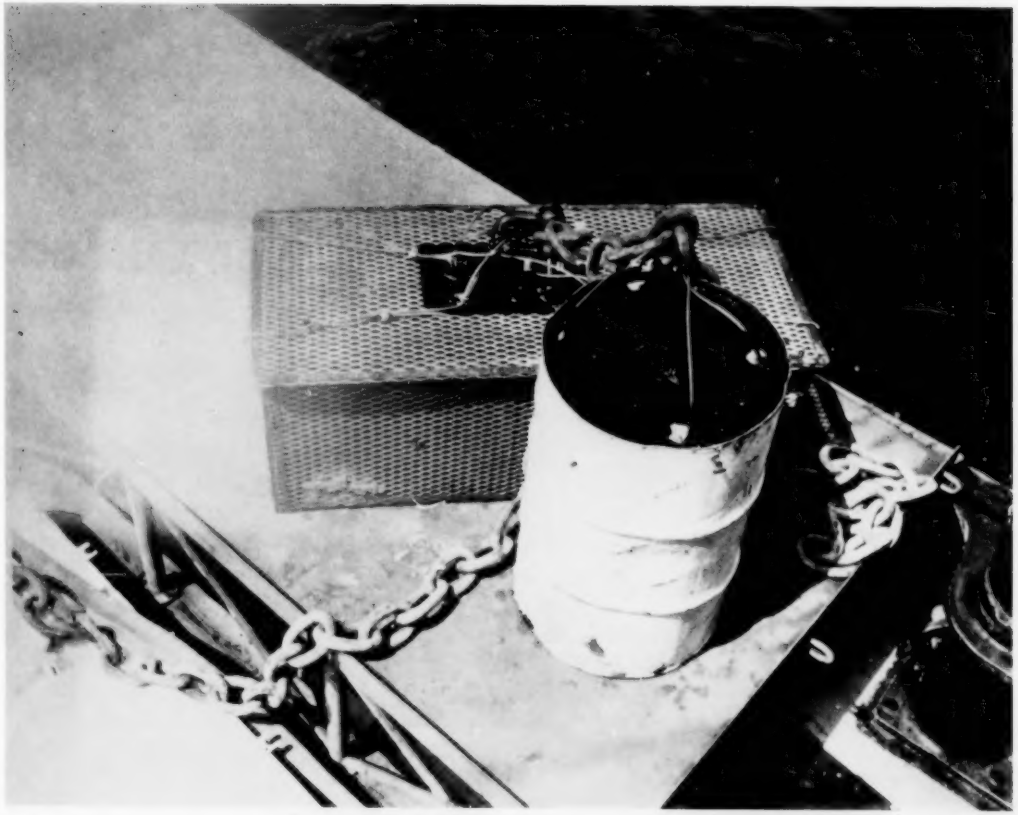

FIG. 1. Vault and drum retrieved after 1 hr at 1025 fathoms. The drum shows a pressure deformation at a filling defect.

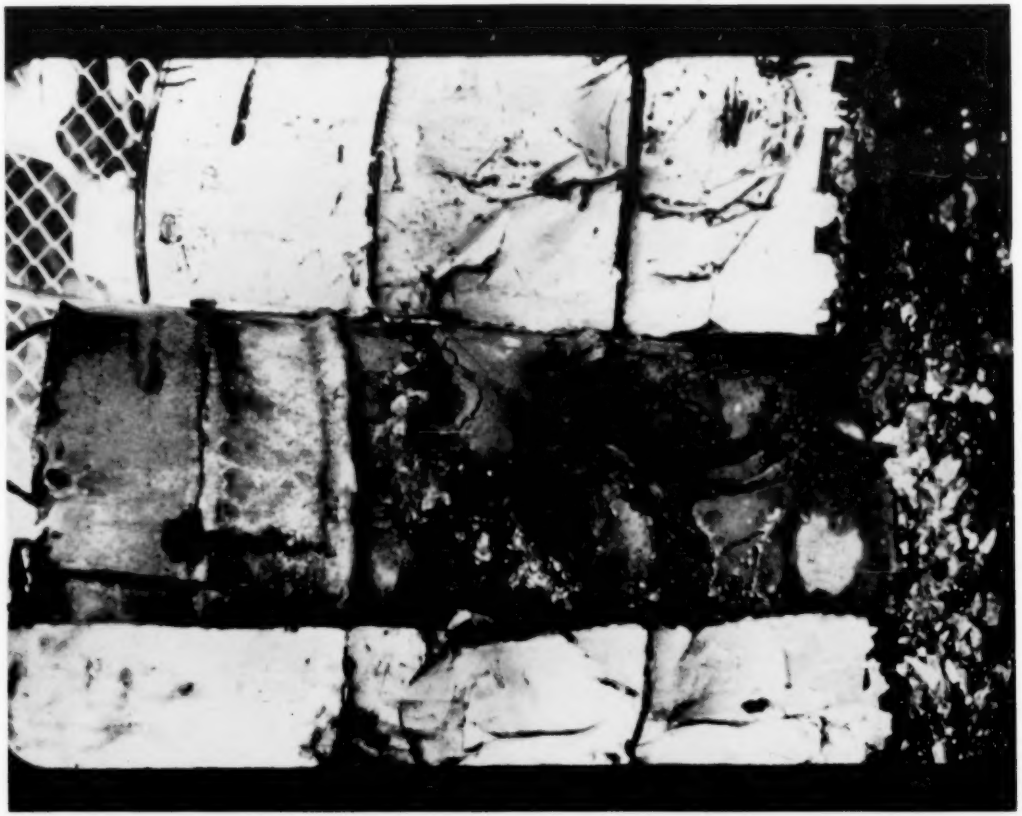

FIG. 2. Recovered drum after removal of steel shell. The steel deformed but did not rupture at each filling defect.

FIG. 3. Paint can and surrounding concrete showing wrinkling from pressure.

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NEWS

Editor: J. J. FITZGERALD, *Associate Editor:* J. C. COLLINS

Regulations and Standards

AEC proposes to amend regulations on radioisotope concentration limits

The Atomic Energy Commission is proposing an amendment to its regulations which will provide additional ways to determine a permissible concentration limit for a mixture of radioisotopes contained in effluents released into air or water.

The Commission amended its regulations, 10 CFR Part 20, "Standards for Protection Against Radiation," on January 1, 1961, to incorporate recommendations of the Federal Radiation Council and the National Committee on Radiation Protection with regard to the total permissible lifetime accumulated dose of radiation. At that time the appendix to the regulation concerning concentrations in air and water above natural background radiation was revised to reflect changes in the basic dose limits. This appendix established concentration limits for certain mixtures of radioisotopes where radionuclides with low concentration limits are not present.

The concentration limits presently specified in the regulation are not sufficiently comprehensive to provide limits for all mixtures of radioisotopes encountered in operations conducted by Commission licensees.

The proposed amendment to Part 20 of AEC regulations would provide an additional means for deriving a concentration limit where (1) the identity of each radioisotope in the mixture is known but the concentration of each radoiisotope is not known; or (2) where the identity of each radioisotope in the mixture is not known, but it can be demonstrated by assay or by the process of elimination that radioisotopes other than those presently specified in the note to Appendix "B" of the regulation are not present to any significant extent.

The concentration limit for any mixture of radioisotopes would be based upon the radioisotope which is or could be present in the mixture and which has the lowest concentration limit now specified in the regulation. The amendment also would establish criteria for determining conditions under which radioisotopes would be considered as not present in the mixture.

Since this method for deriving the concentration limit is based upon the limit for the most hazardous radioisotope that could possibly exist in the mixture, the degree of radiation protection provided by the amendment is at least equal to the radiation protection currently provided under Part 20.

Rules and regulations for the control of radiation under revision in Massachusetts

Both the Department of Public Health and the Department of Labor and Industries in Massachusetts are revising their rules and regulations with respect to the control of radiation hazards and the protection of the employee and the public. The latest amendments of 10CFR20 and the recommendations of NCRPM are being integrated into their final report.

American Standards Association proposes a safety standard for fissionable materials

The ASA N6.8 on Fissionable Materials Outside of Reactors directed by Dixon Callihan, ORNL, has completed a "Safety Standard for Fissionable Materials." It is the expressed intent of this standard to provide guidance in the prevention of nuclear chain reactions in all procedures for handling, storage, processing, transportation and treatment of fissionable isotopes. Generalized basic criteria are presented in the Standard Solutions to specific problems.

Amendments on public records proposed by AEC

The Atomic Energy Commission proposes to amend 10 CFR Parts 9 and 20 of its regulations to include in the public records of AEC all reports of nuclear incidents filed by Commission licensees. The proposed amendments provide that reports of licensee incidents will be available for public inspection in the Commission's Public Document Room.

The Commission believes these reports of licensee incidents should be made available because of the public's interest in information concerning the health and safety aspects of atomic energy activities. In view of the personal nature of the information, the names of persons who have been exposed to radiation will not be made a part of the public records. Separate reports will be made to the Commission of the names of exposed individuals, and these reports will not be made public.

AEC invites comment on proposed agreement with Kentucky for transfer of regulatory authority

The Atomic Energy Commission has republished in the Federal Register for public comment a

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proposed agreement with the Commonwealth of Kentucky for the transfer to the state of certain regulatory authority over radioisotopes, the source materials uranium and thorium, and small quantities of fissionable materials.

Authority which the states may assume under a 1959 amendment to the Atomic Energy Act includes responsibility for rulemaking, licensing, inspection and enforcement in the use of the specified radioactive materials. Under the law, the method of assumption by the states of this regulatory authority is by agreement between the Governor of a state and the Commission. The AEC must make a finding that the state program is compatible with that of the Commission and that it is adequate to protect public health and safety.

Kentucky proposes to assume all regulatory responsibility permitted under the law. This would include authority to regulate the disposal of low-level radioactive wastes other than into the sea or ocean and would also include regulation of the transfer by the manufacturer of products containing atomic material.

The Commission has not taken a position as to whether it should retain, or relinquish to the states, its authority to regulate commercial disposal of radioactive waste by land burial or its authority to license the distribution by manufacturers of products containing radioactive material.

If the Commission decides to retain licensing authority over land burial of radioactive waste, it would include a provision to that effect in a proposed regulation, 10 CFR Part 150. Such a rule would not preclude the state from licensing persons within the state to collect, package and provide temporary storage of atomic waste and to transport such waste for land burial, subject to applicable regulations of Federal agencies having jurisdiction over transportation. A license would still have to be obtained from AEC for the operation of a land burial site. The AEC license would prescribe container specifications, burial sites, precautions to protect against and detect undue movement of buried wastes, and types of wastes acceptable for land burial. This rule would apply only to commercial land burial activities. States would be permitted to license the disposal of certain limited quantities of low-level wastes on the licensee's own land such as is now permitted by the Commission.

Factors, which the Commission will consider in determining whether to relinquish its licensing and regulatory authority over burial of atomic waste involve questions of whether research and development programs of the Commission have progressed sufficiently to permit the establishment of satisfactory criteria for the selection and operation of land burial sites outside Federal regulatory control, whether the requirements for long-term maintenance of burial grounds can best be accomplished by the Federal government or the states and whether waste handling and disposal have such interstate aspects that regulatory control should be continued by the Commission.

If the Commission decides to adopt a regulation retaining authority over the transfer of a product or device by the manufacturer, it would not preclude the state from regulating radiation hazards which might arise during manufacture, transportation or use of such products and devices.

In determining whether to retain this authority over the distribution of products, the Commission must consider, among other things, whether continued control is needed to achieve uniformity of safety design and labeling requirements for such products, many of which are widely distributed; and whether continued Federal control over such products is needed to assure that appropriate limits are maintained on the total quantity of radioactive materials entering the general environment.

Commission revises proposed regulations on shipment of spent fuel elements

The AEC has published for comment procedures and criteria to be followed by AEC licensees to safeguard against accidental criticality, radiation exposure to individuals and the release of fission products during the shipment of spent fuel elements from reactors.

The criteria and procedures apply to solid irradiated fuel elements containing 2000 or more curies of radioactivity in a single shipping container. Smaller shipments will be made as regular radioactive material shipments under existing Federal regulations.

Each licensee will be required to obtain Commission approval of a cask design and shipping procedures. Once approval of a cask design and procedures has been obtained, a licensee may make an unlimited number of shipments using the same casks, subject to the requirements of the Commission's regulations and to the conditions and limitations of his license.

Methods of fastening casks to vehicles or vessels and the operation of vehicles and vessels are not covered in this proposed regulation since these requirements are prescribed by regulations of other Federal agencies having jurisdiction over carriers engaged in interstate commerce. The Inter-Agency Committee on Transportation of Radioactive Materials is examining existing rules governing interstate carriers to insure that those rules are

compatible with the proposed AEC regulation. At present, shipments of irradiated fuel elements require special approval of other Federal regulatory agencies. That practice will continue.

On March 17, 1960, the Commission first published for public comment a proposed regulation on the shipment of spent fuel elements. The new proposed regulation, 10 CFR Part 72, has been prepared in light of comments received, further study by the AEC staff and following an Industry Advisory Conference held May 22, 1961, at which additional comments were received. The new proposed regulation was discussed with the Inter-Agency Committee on Transportation of Radioactive Materials, composed of Federal agencies concerned with the transportation of radioactive materials. An American Standards Association task group has also been consulted.

Federal Radiation Council sets forth recommendations for guidance of federal agencies

The Council has made recommendations for the guidance of federal agencies with respect to environmental radiation levels. These recommendations include:

(1) Radiation protection guides for certain organs of individuals in the general population as well as averages over suitable samples of exposed groups.
(2) Guidance on general principles of control applicable to all radionuclides occurring in the environment.
(3) Specific guidance on exposures to population groups from Ra^{226}, I^{131}, Sr^{90} and Sr^{89}.

Studies by the staff of the Council indicate that observed concentrations of radioactive strontium in food and water do not result in concentrations in the skeleton (and consequently in radiation doses) as large as have been assumed in the past. However, concentrations of I^{131} in the food or water of small children, particularly in milk, equal to those permitted under current standards would lead to radiation doses to the child's thyroid which, in comparison with the general structure of current radiation protection standards, would be too high. This is because current concentration guides for exposure of population groups to radioactive materials in air, food and water have been derived by application of a single fraction to corresponding occupational guides. In the case of I^{131} in milk, consumption of milk and retention of iodine by the child may be at least as great as by the adult, while the relatively small size of the thyroid makes the radiation dose to the thyroid much larger than in the case of the adult. In addition, there is evidence that irradiation of the thyroid involves greater risk to children than to adults.

The Radiation Protection Guides (RPG) recommended below are considered by the Council to represent an appropriate balance between the requirements of health protection and of the beneficial uses of radiation and atomic energy. The following Radiation Protection Guides are recommended by the FRC to be adopted for normal peacetime operations.

The Council also recommended that the radiological health activities of Federal agencies in connection with environmental contamination with radioactive materials be based, within the limits of the agency's statutory responsibilities, on a graded series of appropriate actions related to ranges of intake of radioactive materials by exposed population groups, as stated in Table 2.

The Council recommended guidance on daily intake be adopted for normal peacetime operations to be applied to the average of suitable samples of an exposed population group (Table 3).

In addition, the Council recommended that for radionuclides not considered in this report, agencies use concentration values in air, water or items of food which are consistent with recommended Radiation Protection Guides and the general guidance on intake.

Table 1. *Radiation protection guides for certain body organs in relation to exposure of population groups*

Organ	RPG for individuals	RPG for average of suitable sample of exposed population group
Thyroid	1.5 rems/year	0.5 rem/year
Bone marrow	0.5 rems/year	0.17 rem/year
Bone	1.5 rems/year	0.5 rem/year
Bone (alternative guide)	0.003 μg of Ra^{226} in the adult skeleton, or the biological equivalent of this amount of Ra^{226}	0.001 μg of Ra^{226} in the adult skeleton, or the biological equivalent of this amount of Ra^{226}

Table 2. Graded scales of action

Ranges of transient rates of daily intake	Graded scale of action
Range I	Periodic confirmatory surveillance as necessary
Range II	Quantitative surveillance and routine control
Range III	Evaluation and application of additional control measures as necessary

Table 3. Ranges of transient rates of intake (μc/day) for use in graded scale of actions summarized in Table 2

Radionuclides	Range I	Range II	Range III
Ra^{226}	0–2	2–20	20–200
I^{131}*	0–10	10–100	100–1000
S^{90}	0–20	20–200	200–2000
S^{89}	0–200	200–2000	2000–20,000

* In the case of I^{131}, the suitable sample would include only small children. For adults, the RPG for the thyroid would not be exceeded by rates of intake higher by a factor of 10 than those applicable to small children.

In the future, the Council will direct attention to the development of appropriate radiation protection guidance for those radionuclides for which such consideration appears appropriate or necessary. In particular, the Council will study any radionuclides for which useful applications of radiation or atomic energy require release to the environment of significant amounts of these nuclides. Federal agencies are urged to inform the Council of such situations. Report No. 2, "Background Material for the Development of Radioactive Protection Standards" is available from the Superintendent of Documents, U.S. Printing Office, Washington, D.C. ($0.20).

Federal Radiation News

AEC publishes research pamphlet on atmospheric radioactivity and fallout

The Atomic Energy Commission has published the fourth pamphlet in a series describing its life sciences research program. This booklet, titled "Atmospheric Radioactivity and Fallout Research", summarizes work in progress at thirty-six institutions under forty research contracts or projects.

This booklet reflects the increasing attention now being given to the investigation of atmospheric radioactivity problems associated with areas of the Commission's work, such as the development of nuclear power for missiles and space vehicles.

The broad study areas within which the present research efforts are being conducted are:

(1) Determination of the concentration and physical and chemical characteristics of radioactive fallout and other trace materials in the atmosphere, including analysis of samples collected by balloons, jet aircraft and rockets, as well as by ground stations (atmospheric chemistry).

(2) Theory and observation of the nature of atmospheric motions which transport, mix and deposit radioactive contaminants and tracers (atmospheric dynamics).

(3) Influences of conditions, such as height of burst, at the time of nuclear detonations (whether in air, water or soil) on the characteristics of resulting fallout (fallout formation and source effects).

(4) The relationship of diet, fallout rate and accumulated fallout to observed human body burdens of man-made radioactive materials (radioactivity in soil, food and man).

The publication "Atmospheric Radioactivity and Fallout Research" (TID-12616), may be bought from the Office of Technical Services (OTS), U.S. Department of Commerce, Washington 25, D.C., at $0.75 a copy.

Radiation emergency assistance plan approved by federal agencies

Eleven federal agencies have approved an inter-agency radiological assistance plan to provide advice and assistance upon request from organizations or individuals, in connection with incidents resulting in the loss of control of radioactive materials which involve a hazard or possible hazard to life, health or property.

The inter-agency radiological assistance plan designates the AEC as the agency responsible for the administration of the plan with the co-operation of the other participating federal agencies.

The Interagency Committee on Radiological Assistance, which has been responsible since its formation in May, 1958, for the development of the plan, will continue to function. This committee is made up of officials designated to represent the participating federal agencies.

Under the new plan, federal agencies will co-ordinate their radiological assistance health and safety activities with those of state and local health, police, fire, civil defense and other interested agencies. The plan also calls for setting up a system for the

exchange of information among the national and local groups.

The plan proposes the encouragement of the development of local capability to cope with radiological incidents and the development of basic instructional material in radiation protection that will be made available for public safety and public health personnel.

Reactor News

Radiation levels of SL-1 *reduced by decay and equipment decontamination*

The Atomic Energy Commission announced on September 6 that radiation readings in the SL-1 (Stationary Low Power Reactor No. 1) at the National Reactor Testing Station, Idaho, were reduced to a level of 15 r/hr immediately above the reactor head and 10 r/hr on the operating floor near the reactor. The SL-1 was the reactor involved in a nuclear criticality accident on January 3, 1961.

The readings show a reduction of more than 90 per cent in radiation levels since major decontamination work was started early in June of this year. Readings in the reactor room immediately following the accident ranged from 500 to 1000 r/hr but had decayed to maximums of less than 200 r/hr in April.

Levels of radiation were reduced by manually removing larger pieces of debris, vacuum-cleaner removal of radioactive dust and small particles, and placing bags of lead and steel "shot" around and over the top of the reactor for shielding. The decontamination, dismantling, and analysis work is being carried out by General Electric Company's Idaho Test Station.

Further investigation of the status of the central control rod at the time of the incident indicates that the control-rod extension was "frozen" at a point below the top of the reactor owing to collapse of the control-rod-expansion guide tube. At the time that the control-rod extension froze, the central control rod had been withdrawn three inches but still extended through the reactor core. This seems to cast doubt on earlier theories that the accident could have been induced by rapid and extensive motion of the central control rod.

The disassembly of the reactor vessel may begin within a period of four to six months. At the end of this time, it is estimated that the dose rates would be sufficiently lower to permit the disassembly of the reactor.

Isotope News

International meeting on radioisotopes held in Japan

The Fourth Japan Conference on Radioisotopes was held in Kyoto, Japan, on October 10–12. The purpose of this conference, which was sponsored by the Japan Atomic Industrial Forum, Inc., and the Japan Radioisotopes Association, Inc., was to strengthen the liaison and co-operation between international academic and industrial circles in the field of radioisotopes and radiation through the exchange of scientific and technical information and to make possible wider research and development in the rapidly growing radioisotopes field.

Among those participating were representatives of Australia, Republic of China, Republic of Korea, India, Pakistan, Philippines, Thailand, Canada, France, Czechoslovakia and the United States. Delegates from other Asian and European countries and such organizations as the International Atomic Energy Agency were also in attendance.

The U.S. AEC delegation was lead by Dr. PAUL AEBERSOLD, Director, Division of Isotopes Development, and by Dr. H. D. BRUNER, Assistant Director for Medical and Health Research, Division of Biology and Medicine. Eleven AEC sponsored papers were given at the conference. Approximately 300 papers were presented during the meeting.

Aebersold reports in Japan on the use of radioisotopes

At the Fourth Japan Conference on Radioisotopes, Dr. PAUL AEBERSOLD, USAEC reported the following data.

(1) There are approximately 1900 industrial firms licensed to use radioisotopes.

(2) In the USA, more than 500,000 patients receive tracer amounts of radioisotopes each year for diagnosis of a variety of clinical conditions. More than 100,000 patients annually receive larger amounts of activity for therapeutic purposes.

(3) Approximately 6300 licenses have been issued for research and application of radioisotopes in all fields of endeavour.

(4) The production and use of radioisotopes has grown rapidly. Calcium-47 and I^{131} are being made available on a routine basis. Large sources of Co^{60} and fission products (Sr^{90} and Cs^{137}) are being prepared on a more frequent basis.

American Board of Industrial Hygiene Prepares question on Health Physics for Certification

Under the direction of E. C. BARNES, Westinghouse, a Sub-Committee on the Radiological Aspects of Industrial Hygiene is preparing questions for the certification of Industrial Hygienists.

The Certification Examination will be given in two parts. The first part will consist of a set of questions given to all applicants for certification. The questions in the first part will involve the general

aspects of industrial hygiene, whereas the questions in the second part will require consideration of specific aspects of industrial hygiene. Each part or section will require a day of examination. The health physics examination will be given on the second day for those persons qualified and desirous of being certified as an industrial hygienist with a specialty in the radiological aspects of industrial hygiene.

National Committee on Radiation Protection and Measurements

Handbooks

Publication of three new NCRP–NBS handbooks can be announced at this time. Handbook 75 (NCRP Report No. 25), "Measurement of Absorbed Dose of Neutrons, and of Mixtures of Neutrons and Gamma Rays" was released on February 3, 1961. Handbook 76 (NCRP Report No. 26), "Medical X-ray Protection up to Three Million Volts" was released on February 9, 1961. Handbook 79 (NCRP Report No. 27), "Stopping Powers for Use with Cavity Chambers" was released on September 1, 1961. Also of interest (although not an NCRP handbook) was the release on January 16, 1961, of NBS Handbook 78, "Report of the International Commission on Radiological Units and Measurements (ICRU), 1959". (As in the past, all of these handbooks are available from the Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C.

Handbook 80 (NCRP Report No. 28), "A Manual of Radioactivity Procedures", is now at the printers and should be released in the near future.

The NCRP has received indications that considerable confusion has resulted from the attempts of users of NCRP–NBS handbooks to apply the recommendations of Handbook 76 in conjunction with the advice given in Handbook 50, "X-ray Protection Design" (not an NCRP Handbook). In order to alleviate this confusion, and in view of the fact that the recommendations in Handbook 50 (published in 1952) are now outmoded because of the 1958 changes in maximum permissible dose, WYCKOFF and TAYLOR, who authored Handbook 50, have declared that handbook obsolete.

Subcommittees

Subcommittee 14, on Permissible Exposure Doses Under Emergency Conditions, has completed work on a report directed particularly to the problems of civil defense officials. The report, "Exposure to Radiation in an Emergency", has been reviewed by the Main Committee of the NCRP and is now being prepared for publication.

Subcommittee 6, on Handling of Radioactive Materials, has completed a report, "Safe Handling of Radioactive Isotopes". This report is now undergoing review by the Main Committee.

Subcommittee 11, on Incineration of Radioactive Waste, has prepared a draft of a handbook on this subject which will be submitted to the Main Committee of the NCRP in the near future.

Subcommittee 12, on Electron Protection, is presently revising an earlier draft of a handbook covering this area. This revision is expected to be completed soon and will be submitted to the Main Committee for review at that time.

Subcommittee M-4, on Relative Biological Effectiveness, has continued its active program and is expected to begin writing a report in the near future.

With the death last year of Dr. EBERHARD, the former Chairman of Subcommittee 3, X-ray Protection up to Three Million Volts, and the recent publication of Handbook 76 which had been prepared by that group, it became necessary to reorganize the subcommittee. The reorganization is now complete. ROBERT GORSON has agreed to accept the Chairmanship of the Subcommittee and the following have been named as members of the Subcommittee:

Working Group	*Advisory Group*
R. O. GORSON, Chairman	C. B. BRAESTRUP
J. H. HALE	E. T. FOCHT
J. S. KROHMER	E. R. MILLER
R. R. NEWELL	J. M. MOZLEY
E. D. TROUT	R. J. NELSON
W. J. TUDDENHAM	S. W. SMITH
E. W. WEBSTER	B. P. SONNENBLICK

While the Subcommittee does not anticipate revision of Handbook 76 in the immediate future, a number of questions of interpretation of that handbook have arisen and the Subcommittee expects to deal with these problems as its first order of business.

Reorganization of Subcommittee 7, on Monitoring Methods and Instruments, following the retirement from NCRP activities of the former Chairman, H. L. ANDREWS, is presently underway. A new Chairman, ARTHUR R. KEENE, has been selected, and the following have been named members of the Subcommittee:

A. R. KEENE, Chairman	R. T. MOONEY
J. S. HANDLOSER	J. OVADIA
M. EHRLICH	W. C. ROESCH
J. S. KROHMER	E. W. WEBSTER

It is planned that the Subcommittee will write a new handbook to replace Handbook 51 (NCRP Report No. 10), "Radiological Monitoring Methods and Instruments" which was published in 1952 and hence is somewhat out of date.

It has recently been brought to the attention of the NCRP that the increasing use of radiation-producing equipment in high-school and college classrooms presents a radiation protection problem which has received very little attention up to the present. The Executive Committee of the NCRP has now authorized the establishment of a subcommittee to review this problem with the intent to formulate recommendations on this area. Professor F. SHORE of Queens College has agreed to chair the Subcommittee. The activities of this Subcommittee are to be coordinated with the activities of the Committee on Apparatus for Educational Institutions of the American Association of Physics Teachers which is very much interested in the formulation of recommendations applicable to this area and in the dissemination of such recommendations to the teaching profession. The United States Public Health Service has tentatively agreed to carry out such field studies as may be necessary to obtain some of the operational information needed by the Subcommittee.

Health Physics Activities in England

H. J. DUNSTER

A national policy for emergency exposure

Following any major accident at a nuclear installation, particularly a reactor, there is a possibility that members of the public may be exposed at levels higher than those recommended by the International Commission on Radiological Protection. Such over-exposures are not necessarily harmful because they are transient, whereas the ICRP recommendations refer to continued exposure over a whole lifetime. In many cases remedial action can be taken during the accident to reduce subsequent exposures of the public, and the question arises as to how far this action should be extended. Thus, at the time of the Windscale accident in October, 1957, it was decided that it was not reasonable to allow the dose to the thyroid of children to exceed about 20 rads, if control of consumption of milk was sufficient to keep doses below this level. The choice of an acceptable emergency dose for this purpose and for the assessment of other potential accidents is not easy, and valuable guidance has now been given in the United Kingdom by the Medical Research Council. In three reports, they have now advised on the permissible emergency exposures of members of the public, including children, to certain fission products in diet and breathing air and to external radiation.*

The detailed recommendations include permissible concentrations in milk, one of the principle foodstuffs, and in breathing air, as well as external radiation doses, and the basic body and organ doses from which these figures have been derived are summarized in Table 1.

External radiation control in industry

In a news item in *Health Physics* **4,** 416 (1960), reference was made to draft regulations to control the use of ionizing radiation in factories in the United Kingdom. These regulations have now been issued as a Statutory Instrument and come into full force in February, 1962. They are known as the Ionizing Radiations (Sealed Sources) Regulations, 1961, and can be obtained from H.M. Stationery Office, London. They specify not only permissible radiation doses, but also rules for operating procedures, including working methods, monitoring requirements and medical supervision. A companion set of regulations for unsealed radioactive substances has been published in preliminary draft form, but these regulations are not expected to be operative for some time to come.

A valuable manual for isotope users

Detailed advice on the safe handling of radioactive materials is available in a number of forms, but for the most part such advice is aimed more at the large institution than at the small user dealing with small amounts of radioactivity. The Medical Research Council has now published a revised version of its Introductory Manual on the Control of Health Hazards from Radioactive Materials. This is aimed primarily at the small user and gives valuable advice clearly linked to the degree of hazard of the various types and quantities of radioactive material. It avoids the pitfalls of excessive complexity and excessive caution, but nevertheless sets a pattern which, if followed with care, will provide an exceedingly high standard of safety. The manual, known as MRC Memorandum No. 39, is available, price 1*s.* 9*d.* from H. M. Stationery Office, London. (Approximately $0.25.)

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* Maximum permissible dietary contamination after an accidental release of radioactive material from a nuclear reactor, Appendix J—The hazards to man of nuclear and allied radiations, A Second Report to the Medical Research Council. Cmnd. 1225, H.M.S.O., London (1960).

Report on emergency exposure to external radiation, *Appendix K. loc. cit.*

Maximum permissible contamination of respirable air after an accidental release of radioiodine, radiostrontium and caesium-137, *Brit. Med. J.* Vol. 2, p. 576, August (1961).

Table 1. Maximum permissible organ and body doses for members of the general public following an accidental release at a nuclear installation

Organ	Radiation source	Maximum permissible dose
Thyroid	Iodine isotopes	25 rads (mean over thyroid)
Bone	Sr^{89}	15 rads (local) 3 rads (mean)
	Sr^{90}	1.5 rads/year (local) 0.3 rad/year (mean)
Whole body	Cs^{137}	10 rads
Foetus	External γ-radiation	10 rads
Whole body of children under 16 years and pregnant women	External γ-radiation	20 r (exposure dose)
Whole body except as above	External γ-radiation	30 r (exposure dose)
Superficial tissue of children under 16	External β- + γ-radiation	75 rads
	Skin contamination (10% of surface)	75 rads (additional)
Superficial tissue of adults	External β- + γ-radiation	150 rads
	Skin contamination (10% of surface)	150 rads (additional)

A number of conditions apply to these recommendations and for these and the details of their derivation and of the permissible concentrations, the reader is referred to the original reports.

Health Physics Activities in France

F. Duhamel

OEEC establishes radioactive airborne monitoring and reporting system

The Organization for European Economic Co-operation (OEEC) decided on July 7th, 1961, to establish a monitoring and alert system. This system would operate in a manner such that abnormal increases in the airborne concentrations of radioactive materials would be reported on an international scale. The levels of concern were defined in the manner that is stated below.

The "*Alert Level*" case, wherein the increase of airborne concentrations is such that radiation protection measures are needed.

The "*Pre-alert Level*", wherein the increase is large but not enough to require radiation protection measures.

The "*Scientific Interest Level*", wherein the levels are mainly of scientific interest.

These levels correspond respectively to airborne gross radioactivity of 1000, 100, and <5 $\mu\mu$ c/m^3.

IAEA and Monaco conduct studies on the effects of sea disposal

Under an agreement that has been established between the International Atomic Energy Agency, the Monaco Principality Government and Monaco Oceanographic Institute, studies on the effects of radioactive sea disposal will be carried out by the Agency.

The studies undertaken at the Scientific Center of Monaco will include: a study of the marine currents; measurements of biota; studies of the mineral and biological sedimentary processes; and determination of the background activity distribution and of the accumulation of activity in marine organisms from radioactive waste disposal. In essence, the effects of this disposal on marine ecology will be studied.

With respect to waste disposal operations and studies, a resolution was passed by the French Academy of Medicine in favor of recommendations that were made by Professor Tanon. The following recommendations were made:

Experiments conducted by biologists and oceanographers, on the behaviour of fauna and flora,

especially plankton, in water contaminated by radioactive wastes, should be pursued.

Disposal should be made only after approval has been given by the official oceanographic authorities. Health authorities should be informed of those studies and of the results of these studies.

The Academy also expressed a desire for an international agreement on waste disposal and for the judicious selection of land storage sites that all countries might use.

Radioprotection Congress to be held at Orsay

At Orsay, France, a Radioprotection Congress will be held during the fall of 1961 or the Winter of 1962. The Congress has been organized under the direction of Mr. PAUL BONET-MAURY at the Nuclear Physics Laboratories of the Radium Institute in Orsay, France. The Radio Protection Society of France, a society composed of French technicians engaged in radiation protection activities will be assisted by the French Section of the Health Physics Society in the conduction of the meeting which will principally be concerned with the shielding problems of high-energy accelerators. In France, studies are presently being made on a 15-BeV, 5-A accelerator project. An alternative design of the above project was presented by Mr. BRUCH at the Brookhaven Congress, (September 5–12, 1961).

French research reactor in operation

Criticality was achieved in June 1961 in Ulysse Research Reactor. Formal start-up has been made by Mr. JEAN DEBIESSE, Saclay Nuclear Research Center and Nuclear Sciences and Techniques National Institute Director. Ulysse is a reactor of the Argonaut family, water moderated, air and water cooled. Fuel consists of 90 per cent enriched uranium and core critical mass is 50 kg of U^{235}. Core mean thermal flux is about 10^{12} n/cm^2/sec. Ulysse is the prototype of a low-cost reactor family designed for training and nuclear research centers equipment.

Cadarache Nuclear Center Site progress is reported

At the Cadarache Nuclear Center Site, progress is being made on the Pegase, Pegase Mock up (Peggy) and Rapsodie reactors. Enriched uranium processing, and plutonium purification facilities are being built.

Construction work on Rapsodie began in July; Rapsodie will be the first sodium-cooled fast breeder in France. The fuel will consist of a Pu–U–Mo or PuO_2–UO alloy. Critical mass is expected to be 50 kg Pu^{239} and 80 kg of U^{235}. Power level will be 8.5 MW (thermal). The center-core fast flux will be 1.1 $\times$ 10^{15} n/cm^2/sec. Rapsodie should be in operation in 1963.

A series of experiments is presently being carried out on the site. Environmental atmospheric radioactivity monitoring stations are being equipped. A large-scale atmospheric diffusion experiment, the so-called "Bee Operation", has provided fallout contours at ranges of 1, 2, 3 and 4 km. Monitoring Stations of the Durance River will be provided with continuous and automatic detectors, built by CEA. Preliminary studies have been made on the treatment of Pegase effluents.

Health Physics Activities in Italy

ENRICO VIA

Italian Health Physics Association meets in Turin

The Seventh Meeting of the Italian Health Physics Association took place in Turin on June 3–6, as part of the International Medical and Surgical Meeting.

The Health Physics Association discussions, at which the French scientists were present, dealt with:

(1) Measuring the radioactivity in water.

(2) Control of radioactivity in the alimentary chain.

(3) The clinical and laboratory criteria for the selection of workers in the nuclear industry.

At the end of the meeting, the participants paid a visit to the Nuclear Research Centre of the Societa Ricerche Impianti Nucleari (SORIN) at Saluggia, near Vercelli.

Effects of sea disposal under study in Italy

Since 1959, a laboratory for the study of problems that are caused by radioactive contamination of the sea has been in operation at Fiascherino (Province of LaSpezia). The program of this laboratory includes a determination of the factors that influence the absorption of radioisotopes by marine organisms, and the passage of radioactive substances in the sea through feeding chains to those fish products which are ultimately used for human consumption.